

# Feasibility Study of a Proliferation Resistant Fuel Cycle for LWR-Based Transmutation of Transuranics

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## 1.0 Abstract

*Options for transuranics recycle in PWRs with the CORAIL concept have been assessed assuming different separation hypotheses. The cases evaluated include those with Pu-only recycle and transuranics (TRU) recycle, in addition to others investigating partial recycle of the transuranics (e.g., Pu+Np, Pu+Np+Am, and fractional recycle of Pu and Am plus Np). The impacts of these options on proliferation resistance, fuel handling and the repository were evaluated. The results indicate that the option of all TRU recycle is the one most beneficial to the repository and non-proliferation. The case however results in the most problems for fuel handling at the fabrication stage as a result of the high spontaneous fission neutron emission rates. Pu+Np multirecycling offers marginal additional benefits over Pu-only multirecycling, as it does not give significant increases in the radiation dose and behaves similarly to the Pu-only spent fuel in the repository environment. Recycling americium in the fuel cycle provides significant benefits to both non-proliferation and the repository, over that obtained with Pu-only recycling. Coupling americium recycling in the fuel cycle with storage of curium is an option that appears promising, if an appropriate solution for curium storage is found.*

*Since TRU multirecycling in PWRs is attractive, because of existing technology for MOX fuel and the potential cost advantages over dedicated TRU burners, additional evaluation of the option was performed to assess the number of recycle stages that are practical before fuel handling considerations result in preventive measures that make the fuel cycle expensive or impractical. Limited proliferation evaluations were also performed for the option. Results for the various performance indices suggest that at least seven recycles can be performed using the CORAIL-TRU concept, provided remote handling of fuel is a price that is acceptable for the transmutation mission. Three recycles with extended cooling interval prior to loading in advanced reactor systems is another option that was proposed. The delay time provided by this campaign could be used for developing and deploying the advanced systems. Even for this option, fuels development and infrastructural needs and shielding requirements necessitate modifications to currently existing PWR fuel cycles.*

## 2.0 Introduction

The scenarios that have been proposed for sustainable nuclear programs have typically included strategies for managing long-lived nuclear waste, as this is considered as one of the issues to be addressed to facilitate future nuclear development. The approaches employed in the FY2001 activities of the USDOE Advanced Accelerator Applications (AAA) program envisioned multi-tier systems in which plutonium (Pu) or transuranics (TRU) in spent nuclear fuel (SNF) from commercial nuclear reactors are first burned in advanced thermal or fast systems and are subsequently burned in fast-spectrum (critical or accelerator-driven transmutation of waste (ATW)) systems.[1] The task of completely burning (finishing off) the TRU was assigned to these latter systems in which the neutron spectrum is considered favorable for efficiently performing the task. The multi-tier approaches were designed to reduce the inventory of nuclear waste that would need to be handled by the ATW systems, thereby reducing the cost and scope of the ATW.

The advanced light-water reactor (ALWR) systems evaluated in the FY2001 multi-tier study assumed full-core loading of mixed oxide (MOX) fuel assemblies or inert-matrix, non-fertile fuel (NFF) fuel assemblies. [1] The role of LWRs in the overall transmutation mission, however, was limited because with one or two recycles of Pu or TRU in such cores, the transuranics content becomes high and results in a positive void coefficient. This is undesirable for reactor safety, and limits the number of recycle stages and hence fuel consumption level in the LWRs. As a result, in the FY2001 study, it was necessary to pass spent transuranics to a second tier fast spectrum reactor or accelerator-driven system in order to complete the transmutation mission. There are clear benefits if TRU transmutation could be performed *completely* in LWR systems or using assembly designs that are retrofittable in current LWR core designs. The perception is that existing technology for Pu recycling would accelerate the commercialization of this approach.

Some U.S. and international studies are addressing the feasibility of this approach using both homogeneous and heterogeneous fuel pin and assembly designs. Options under consideration have included variants of MOX, non-fertile fuels, and thorium based fuels. The focus of this study is the evaluation of MOX concepts for LWR-based transmutation of nuclear waste in a sustainable nuclear enterprise. MIT and Brookhaven National Laboratory (BNL), as part of this USDOE project, are evaluating the options with non-fertile fuel and thorium-based fuel, respectively.

A proliferation resistant fuel cycle is being evaluated for currently operating or evolutionary light water reactors (LWRs). This fuel cycle would permit the burning of either plutonium (Pu) or transuranics (Pu, neptunium (Np), americium (Am), and curium (Cm)) in spent nuclear fuel. In the U.S., the choice between plutonium and transuranics (TRU) would probably not be wholly based on technical considerations, but would require input from the government because of the current restrictions on using Pu-only fuel forms in the commercial fuel cycle, due to non-proliferation concerns. If Pu-only fuel cycles are proscribed for LWRs, then the options for burning the transuranics include using homogeneous plutonium and minor actinides (Np, Am, Cm, and higher actinides) fuel forms or plutonium with a subset of the minor actinides (MA) or plutonium with fractional amounts of the MA. Another option that has been discussed is to add fission products to the fuel to enhance its intrinsic proliferation property, by increasing the radiation dose.

The current work is a preliminary evaluation of various fuels separation hypotheses that have the potential to advance the transmutation goals of the USDOE Advanced Fuel Cycle Initiatives. Partial transuranic (TRU) separation cases and some isotopic separation cases are included in the current study in order to assess which of the options have promise in providing benefits to the repository load reduction and the non-proliferation policy of the U.S. In this regard, fuel cycle indices that can be used to measure the proliferation resistance of the fuel material and assembly and the ease of handling them in the fuel cycle, have been evaluated. The indices investigated include critical mass and radiation sources for the plutonium or TRU in the fabricated fuel for non-proliferation considerations, and decay heat, neutron sources and gamma sources and doses from the fuel at the separation and fabrication stages in the fuel cycle, as a measure of fuel handling issues. Decay heat and radiotoxicity values at the repository were used to provide indications of what the options offer for repository load reduction. The CORAIL assembly concept that has been evaluated under the AAA Downselection Studies [2,3,4] is the focus of this work. Other fuel cycles can be envisioned for the current task, and in fact MIT (non-fertile fuel concepts) and BNL (thorium-based fuels) are studying some potential LWR transmutations systems as part of this project. [5]

In Section 3.0, the evaluation objectives are described. Section 4.0 contains a brief description of the CORAIL assembly concept for plutonium or TRU transmutation. This concept has been extended for partial TRU multirecycling in this current work. The calculation methods employed in the study for evaluating the indices are discussed in Section 5.0. These include using the MCNP code [6] for evaluating the critical mass and

dose rates, and a combined WIMS8 [7] (or TRANSEQM) and ORIGEN2 [8] calculation path for evaluating material decay heat and radiation sources. The findings from the evaluation of various separation hypotheses are presented in Section 6.0. The evaluation of fuel handling issues in the CORAIL-TRU fuel cycle, in order to determine a practical limit to the recycle stages, is presented in Section 7.0. Results obtained for the non-proliferation studies are summarized in Section 8.0. The conclusions from the work and discussion of future activities are contained in Section 9.0.

### 3.0 Evaluation Objectives

Preliminary evaluations of the CORAIL concept for transmutation of transuranics (CORAIL-TRU) have indicated that the concept might be feasible from a neutronic viewpoint, since with limited recycling, the reactivity balance, reactivity coefficients, shutdown margins, and power peaking factor of the traditional UOX or MOX fuel can be preserved.[3] However, questions arise because of the increasing minor actinide content with recycle stage, which tend to make fuel handling more difficult and potentially expensive. The objective of this study is to provide answers to the following questions and issues:

- What other separation hypothesis can be employed in the LWR proliferation resistant fuel cycle, in order to provide benefits to the repository and to minimize fuel handling issues? The possible combinations that can be postulated include: Pu only (as a reference for comparison), Pu+Np, Pu+Am, Pu+Cm, Pu+Np+Am, Pu+Np+Cm, Pu+Cm+Am, and Pu plus fractions of these elements. Others could include the addition of highly radioactive fission products in the fuel mix.
- How many recycle stages are possible in the CORAIL-TRU concept before the fuel becomes impractical to handle?

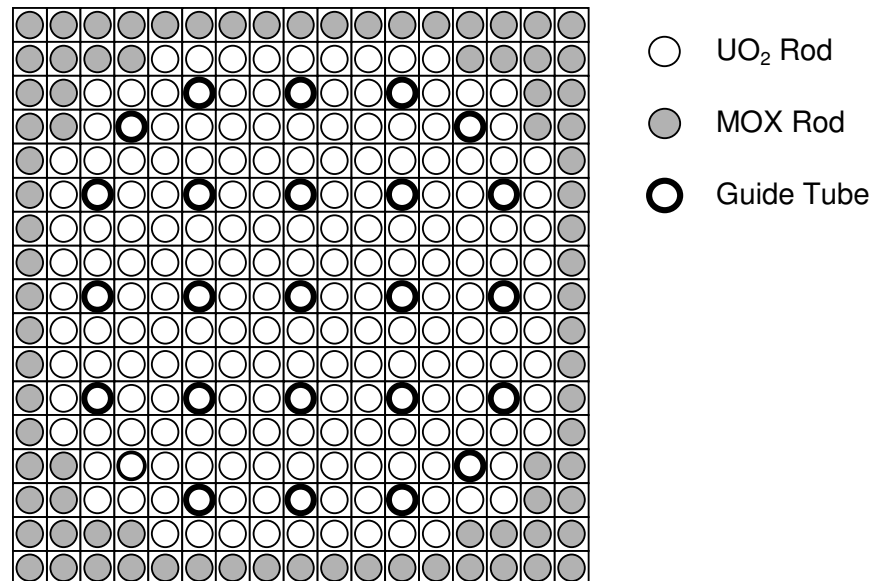
The study of different separation hypotheses was focused on approaches that could be used for minimizing fuel handling problems, arising primarily from the recycling of curium. This restricted the options to a few cases: Pu-only, Pu+Np, Pu+Np+Am; and fractional separation of Pu and/or Am plus Np. The fractional separation cases demonstrate the benefits to the fuel handling consequences when the amount of curium to be handled is minimized. However, in this case, a solution must be found to the stored curium. It will be shown in Section 6.4 that passing the curium in spent fuel to the repository will make it difficult to meet the goal of attaining a radiotoxicity level that is lower than that of the source uranium in a 1,000-year time frame.

The study of the optimum recycle stages in the CORAIL-TRU concept is predicated on the view that a few recycles could be beneficial to the transmutation mission. If two or three recycle stages of CORAIL-TRU are permitted for example, this would result in a twenty to thirty years of operational period over which the TRU inventory in the U.S. nuclear industry could be held constant. This period could be used for further (parallel) development of more advanced reactor systems and fuel cycle concepts. The mass reduction and radiotoxicity issues typically required for repository

benefit analysis are not pertinent to the feasibility of the CORAIL approach in the short-term (2015 or so time frame). However, ultimate solution must be considered, therefore, these additional issues are covered under the section on separation hypotheses (Section 6.0), as a measure of the impact of these scenarios on the repository.

#### 4.0 CORAIL Assembly Concept

This work focuses on the CORAIL assembly concept that has been studied for the AAA Downselection Studies [2,3,4]. Because detailed information on this concept has been provided in Ref. 2 and 3, only a brief description of it is given here for completeness. The heterogeneous CORAIL assembly is designed for multirecycling of plutonium (Pu) or transuranics (TRU) in retrofittable LWR assemblies that can readily replace those currently used in the LWR industry. The assembly design employs both enriched uranium oxide (UO<sub>2</sub>) pins and mixed oxide (MOX) pins (with Pu or TRU) for this purpose. The fuel types have been developed and used extensively in the nuclear industries over the world, although the use of MOX fuel is not as extensive as UO<sub>2</sub> fuel. The enriched uranium fuel is employed for supporting the recycling campaign by limiting both the MOX-pin power and the number of MOX pins in the assembly, to retain conventional core performance. About 30% of the fuel pins are MOX pins, which are placed at the periphery of the assembly. The CORAIL assembly is displayed in Fig. 4.1. Assembly data are presented in Appendix A.



**Fig. 4.1. Pin Loading Pattern of CORAIL Assembly**

The plutonium or TRU is multirecycled in the CORAIL concept with the aim of stabilizing the material. Stabilization implies that the content of plutonium or TRU initially loaded into the assembly is constant over the recycle stages. The CORAIL assembly concept can therefore be envisioned as a “delay-line” for the plutonium or TRU,

as it slows down the accumulation of these hazardous materials in the waste destined for the repository. Following discharge of the assembly, the  $\text{UO}_2$  and MOX pins are co-processed to recover the plutonium or TRU in the assembly. This material is then used for fabricating the fuel for the next stage of the multirecycle operation. Each recycle stage takes about 11.5 years in the CORAIL design being evaluated. Of this time, the fuel is resident in the core for 4.5 years. A 5-year cooling time is assumed after which fuel separation and fabrication take place. An additional 2 years is assumed before the fuel is loaded back into the reactor core. Each operational cycle is 1.5 years and the fuel accumulates 15 GWd/t of burnup in this period. This results in a discharge burnup of 45 GWd/t for the CORAIL assembly.

In the CORAIL-Pu concept, in which the assembly would be used to recycle and stabilize only the plutonium in spent nuclear fuel (SNF), the minor actinides (MA) arising from the campaigns would have to be disposed or burned in a subsequent system. While disposal to the repository would provide mass and radiotoxicity reductions relative to the traditional  $\text{UO}_2$  spent nuclear fuel (SNF), it would not meet the repository performance goals that were identified in FY2001 systems studies. [1] For that work, it was required that the radiotoxicity be reduced to natural uranium levels in less than a thousand years. Alternatively to direct disposal of the MA, the material could be used as fuel for a Tier 2 fast spectrum system that is designed to complete the mission of burning the material. In this case, only losses in the reprocessing stages are passed to the repository and the FY2001 transmutation goals would be readily met. This option has been evaluated as part of the Downselection Studies of the AAA Program [4], and the results indicate that an equilibrium cycle could be sustained with the minor actinides. A creative solution is however required for starting the recycling campaign. In the earlier study, it was suggested that spiking the initial fuel with fissile plutonium from the commercial spent nuclear fuel stockpile might be a preferred approach.

It has also been proposed to stabilize TRU with the CORAIL concept (CORAIL-TRU). Assessment of this approach has also been done under the Downselection Studies [3]. In this case, only the TRU lost during reprocessing passes to the repository. However, questions arise about the difficulty of handling the material because of the increase in minor actinide content with each recycle stage, which makes the material hotter and more radioactive. A preliminary evaluation of some of these fuel handling issues is undertaken in this work (see Section 7.0). Alternative separation options for alleviating these problems are also evaluated.

## 5.0 Calculation Methods

The results employed in this study were obtained using the WIMS8, [7] ORIGEN2, [8] and MCNP4 [6] codes. These are standard codes that are routinely used in the nuclear industry. The WIMS8 lattice code is employed for calculating the core reactivity balance (cycle length) using the linear reactivity model for a three-batch core, and the assembly power peaking factor. The code is also used for estimating the reactivity coefficients for the core. The depletion module of the WIMS8 code tracks heavy nuclides ranging from Th-232 to Cm-245. Additionally, the time evolutions for the masses of over a hundred fission products are also calculated. These nuclides are however only those that have been determined to be pertinent to the neutronics reactivity balance. Some nuclides that are required for heating and radiation sources calculations have therefore been neglected. Because of these considerations, ORIGEN2 calculations are also performed to provide estimate of the nuclide masses for the neglected nuclides. A coupled WIMS8-ORIGEN2 procedure was developed for merging the data from the two codes to give a consistent set of masses for a given problem. This procedure has been described in two CORAIL reports [2,3] and so would not be repeated here.

In addition to the WIMS8-ORIGEN2 procedure, an analytical approach to evaluate directly the equilibrium state of the various CORAIL concepts employed in this study has been developed. The approach is based on a one-group depletion code called TRANSEQM (see Appendix C). This code tracks heavy-metal nuclides from Th-232 to Es-253; it does not track fission products, however. The code is important because using the WIMS8-ORIGEN2 procedure is very cumbersome and time consuming, especially for the CORAIL-TRU cases that require many recycle stages (more than 20) to attain equilibrium. Additionally, that procedure was not tracking accurately the higher actinides. The TRANSEQM code has been verified by comparing the results to those generated with the WIMS8 and ORIGEN-RA [9] codes.

The TRANSEQM code results have also been systematically analyzed to ensure that numerical round-off, transmutation chain approximations, and the assumption of constant neutron flux and cross sections during the irradiation cycle do not result in large errors for the masses of higher actinides (i.e., beyond Cm-245). The verification activity demonstrates that the transmutation chain and the numerical solution of the one-group transmutation code are as accurate as those for the ORIGEN-RA code. A primary assumption in the code is the cross sections used for the higher actinides (beyond Cm-245); those for lower actinides come from WIMS8 calculations. Four group cross sections for the higher actinides are obtained from Ref. 10. One-group cross sections are

then derived by condensing the four group data with WIMS8 generated group fluxes. This assumption is expected to introduce uncertainty in the masses of the higher nuclides, which would affect mostly the neutron source values; these higher actinides are important for the neutron source calculation even though the nuclide masses are low. A preliminary estimation of this uncertainty indicated that the masses could be off by 50% and that this could affect the prediction of the neutron source by this amount. (This estimation was made by evaluating the error that would have been introduced in the cross sections values for which WIMS8 data is available (i.e., Cm-245 and below), if the same approximation is used for them. This is not necessarily representative of the error in the higher actinide (above Cm-245) cross sections because the self-shielding factors for these nuclides should be smaller since most of them exist in small amounts). In any event, additional estimation of the impact of these cross sections is still required. Finally, results from the TRANSEQM code are coupled with those from the ORIGEN2 code, similarly to the approach described for the WIMS8-ORIGEN2 procedure discussed above.

The MCNP4 code was used for calculating the critical masses, and neutron and gamma doses used in this study. These calculations were done for separation products, fuel pellets, and fuel pins. For these calculations, the nuclide masses and neutron and gamma sources obtained from the WIMS8-ORIGEN2 or TRANSEQM-ORIGEN2 procedure are used. With the given masses and source distributions, the MCNP4 code calculates the gamma or neutron flux at the spatial zone boundary of interest. In addition, the gamma flux and neutron flux to dose conversion factors specified in the MCNP4 users' manual (ANSI/ANS(1977)) are then multiplied by the fluxes to obtain the doses. The MCNP4 models used for dose calculations recognize the fact that the spontaneous fission neutrons and neutrons from ( $\alpha$ ,n) interactions have different spectra, because of the different energy phenomena associated with the processes. Additionally, gamma source spectrum obtained from ORIGEN calculations are used in the MCNP calculations.

For the MCNP calculations, the gammas or neutrons are distributed uniformly over the volume of the fuel pellet or pin, or fuel sphere. A vacuum boundary condition is used for all the cases. In the fuel pin case, the pin is assumed clad with Zircaloy-4, and the full length of the pin is modeled; a single cylinder is used to model the fuel. The fuel pellet, pin, and sphere dose rates presented in Sections 6.0 and 7.0 were obtained using this approach.

For the critical mass calculation, it was generally assumed that the transuranics exist in an isolated form, though some cases in which transuranics are mixed with uranium were also evaluated. This calculation was performed for a bare sphere of the material of interest.

## 6.0 Physics Evaluation of Different Separation Hypotheses

Different separation hypotheses have been evaluated in order to investigate their impacts on fuel handling indices and the repository. For these cases, the equilibrium cycle nuclide masses were used in generating the radiation sources at various fuel cycle stages. The different CORAIL multirecycling cases are:

- Pu only multirecycling,
- Pu and minor actinide (i.e., TRU) multirecycling,
- Pu, Np multirecycling,
- Pu, Np, Am multirecycling,
- Pu, Np, Am multirecycling, without Pu-242,
- Pu, Np, Am multirecycling, without Pu-242 and Am-243.

For the latter two cases, it is assumed that the Pu-242 and Am-243 are removed selectively by isotopic separation. These cases were included to provide indications of the relief that is obtained if the presence of curium is minimized in the fuel cycle and the benefit to the repository.

The evaluations in this study were done for the equilibrium states of the different separation hypotheses. The TRANSEQM code was used for searching the equilibrium mass fractions. All the calculations were done with a cooling period of five years between discharge and separation/fabrication, with the exception of one case. For this latter (CORAIL-TRU) case, a cooling period of 20 years was employed. The case was included to assess the impact of allowing Cf-252 to decay over seven half-lives and the Cm-242 to decay over one half-life, on the fuel handling indices.

Table 6.1 is a comparison of the charge TRU compositions at the equilibrium states; the TRU vector is mixed with depleted uranium to form the MOX-pin heavy metal compositions. The resulting enrichment for the UO<sub>2</sub> pins and the TRU content of the MOX pins are provided in the Table. The data indicate that the required TRU content in the MOX pins are less than 10%, except for the CORAIL cases involving additionally the recycle of americium and curium, and higher actinides. In these cases TRU contents as high as 20% are obtained. The highest uranium enrichment requirement is 5.5%, which is higher than that generally available from enrichment services plants.

As more of the higher actinides are included in the multirecycle concept, the TRU content increases because the relative fissile content is decreased and most of the

additional nuclides are absorbers in the thermal spectrum of a PWR. This has impacts on the system reactivity coefficients and power peaking factor. Evaluations performed in the CORAIL-TRU study [3] however indicate that these impacts do not adversely affect the core performance, compared to the  $\text{UO}_2$  assembly case.

No additional evaluation of reactivity coefficients and safety parameters were performed in this study because similar evaluations were done in the previous systems studies for the CORAIL-Pu [2] and CORAIL-TRU [3] concepts. From physics reasoning and from earlier studies [11], it is expected that the core physics parameters of the other partial recycling cases would be between those for CORAIL-TRU and CORAIL-Pu assemblies. The earlier physics studies indicated that the core cycle length requirements can be met with multirecycling of plutonium or TRU in the CORAIL assembly. Reactivity parameters were also found to be similar to those for same size (same MWt-yr)  $\text{UO}_2$  assembly (see Appendix J), because the assembly contains about 70%  $\text{UO}_2$  pins. Importantly, the void coefficient was found to be negative for the assemblies. However, detailed core transient analysis has not been performed, and could be pursued in the future. Additionally, for the CORAIL-TRU core, power peaking could be a problem with increasing plutonium content, as is the case with recycle stages. In Ref. 3, approaches for restricting this problem were discussed.

Discussions of results for the different separation hypotheses are presented in the following sections. The fuel handling indices of gamma and neutron sources, and decay heat, are compared in Table 6.2, for the different scenarios. In the study of the impact of separation hypotheses on the repository, it is assumed that the nuclides that are not recycled are passed to the repository; recycled material remains in the fuel cycle, and only fraction of it is lost to waste during the separation stage. Results for the different cases are summarized in Figs. 6.1 to 6.4, which display the radiotoxicity (normalized to that for the source uranium used in producing the CORAIL fuel assembly) and decay heat values, for the different cases.

### **6.1 Pu-Only Separation**

The separation of plutonium from the legacy LWR spent nuclear fuel appears to be a first logical step in the transmutation mission. Plutonium represents the largest mass component (>85%) of the TRU in spent nuclear fuel and the technologies for recycling plutonium in LWRs are being demonstrated in the countries (e.g., France) that employ it in the nuclear fuel cycle. The retrofittable CORAIL concept was designed to benefit from

this experience. In the U.S. situation, however, the pertinent infrastructure would have to be developed and built prior to the implementation of this scenario.

The burning of plutonium has benefits to the repository. The first is that plutonium would not be stored in the repository, and hence the concern that the repository could be a plutonium mine is removed. Assuming no additional transmutation in a fast spectrum system, the multirecycling of just the plutonium results in a factor of 2 to 5 reduction in the long-term radiotoxicity, compared to the  $\text{UO}_2$  case (see Fig. 6.1). With this scheme, however, the goal of reducing the radiotoxicity to less than that of the source uranium in a thousand years, [1] is unattainable; geologic disposal of minor actinides keeps the radiotoxicity above that of natural uranium ore for at least 50,000 years after disposal.

Long-term decay heat of the discharged fuel is also reduced by recycling the plutonium in the fuel cycle, as indicated by Fig. 6.3 (for the case in which the minor actinides are assumed discharged to the repository). Evaluations have shown that for the same energy production, the mass of minor actinides discharged from the CORAIL-Pu approach is a factor of three greater than that in a  $\text{UO}_2$  assembly [2]. The higher Cm-244 content in the discharge fuel is the reason for the initially higher decay heat value for the CORAIL-Pu case compared to the  $\text{UO}_2$  case (see Fig. H.4 in Appendix H). Because Cm-244 dies away very quickly ( $T_{1/2}=18\text{years}$ ) in the repository setting, the decay heat arising from burying the TRU (minor actinides and plutonium separation losses) in the CORAIL-Pu case then becomes lower in less than fifty years. The particular benefits to the repository in terms of the reduction in the required capacity would have to be evaluated by detailed repository performance assessment. Clearly, something has to be done to the Am-241 in the repository, as it is the leading contributor to the decay heat and radiotoxicity in the 100 to 1,000 year period (see Figs. H.2 and H.4).

The mass reduction provided by plutonium recycling is also an advantage to a multi-tier approach in which fast spectrum systems are employed for completing the mission of TRU burning. In this case, the support ratio is increased, as the overall mass of the TRU to be burned in the latter system would have been reduced by a factor of about 4.

Previous CORAIL studies [2] have indicated that the fuel handling indices for the charged CORAIL assembly with plutonium recycling are higher than those for the  $\text{UO}_2$  fuel, but are not considered problematic; MOX assemblies fabricated with reactor-grade Pu, which contain roughly three times more plutonium than the CORAIL assembly, are

currently utilized in the French nuclear program. Fuel handling concerns are also much lower than for the TRU recycling case.

Finally, the U.S. non-proliferation policy might preclude the option of Pu-only separation. It is however clear that even if the option is permitted, additional safeguards might be imposed on the nuclear fuel cycle. Table 6.3 shows that the radiation dose from the MOX pellet can be reduced significantly by the use of zircaloy cladding (changes from 5 rem/hr to 0.1 rem/hr). This is because relatively low energy gammas (photons) contribute about 99% of the dose. The dose rates on Table 6.3 are judged adequate for the current study because the results (magnitude) for the CORAIL-Pu case has been compared to that found in open literature. According to Ref. 12 (page 73), the pellet surface gamma dose for MOX fuel (4.3% PuO<sub>2</sub>) that has been cooled for 600 days is about 2.3 rem/hr. This compares quite well to the 5 rem/hr shown in Table 6.3, for a MOX fuel with 8.2% PuO<sub>2</sub>. According to the reference, at 61 cm from the surface of an all-plutonium PWR assembly using MOX fuel, the gamma dose rate is about 8 mrem/hr. This trend confirms the greatly reduced values at a distance of one meter away from the fuel pin, presented in Table 6.3. The specific information from Ref. 12 is provided in Appendix I.

## 6.2 TRU (Pu+MA) Separation

The ultimate goal of the transmutation mission is the removal of the problematic components (TRU and long-lived fission products) of nuclear waste. For this reason a separation hypothesis involving the complete separation and transmutation of TRU in the CORAIL assembly was included in this study. As expected, the most benefit to the repository is obtained when all the TRU are multirecycled in the CORAIL assembly. The radiotoxicity and long-term decay heat in the repository, are significantly reduced as indicated by Figs. 6.1 and 6.3, respectively, compared to the UO<sub>2</sub> case. Dose rates to inhabitants in close proximity to the repository are also expected to be significantly reduced; this calculation was not done in the current study, but the elimination of the plutonium, neptunium, and americium ensures that this is the case. The short-term decay heat is not significantly reduced for this case because it is dominated by fission products. Larger reductions (compared to the Pu-only and UO<sub>2</sub> cases) are clearly evident after a few hundred years.

If transuranics could be completely burned in LWRs, a significant reduction in the mass inventory of high level radioactive waste sent to the repository would be obtained. Comparative studies have shown that in this case, the waste radiotoxicity source term of a

multi-recycle PWR would be similar to that of a fast system – the driving factor is the separation effectiveness (assumed to be 0.1% in most systems analysis) [13].

The generally elevated radiation sources and decay heat resulting from TRU recycling is an item that would be new to the U.S. light water reactor fuel cycle. For this reason an evaluation of this effect is included here. In general, the presence of neptunium, americium, and curium in the fuel cycle increases the decay heat and radiation sources, compared to the case with Pu-only multirecycling (see Table 6.2). Recycling the transuranics increases the decay heat value of the charged CORAIL assembly by nearly a factor of 18, and the neutron source by a factor of 38000, at the charge state, relative to the CORAIL-Pu case. Note that these results are for the equilibrium state in which the minor actinide content is high. The results in Section 7.0, for a few recycles, show lower differences between the CORAIL-TRU and CORAIL-Pu cases. The increased decay heat is due to the higher loading of Cm-244 and Pu-238 in the CORAIL-TRU assembly. Additionally, Cm-244 has a relatively short half-life (18 years) and a small, but significant branching ratio for spontaneous fission. The dominant neutron source for the equilibrium cycle of the CORAIL-TRU case is however Cf-252.

The elevated decay heat would preclude contact handling. The higher neutron source would require that additional measures (e.g., shielding and remote fuel handling) be implemented at the reactor to protect refueling operators. An assessment of the neutron doses arising for this assembly concept is presented below and in Section 7.0.

All assemblies have essentially the same total decay heat load (similar to that of  $\text{UO}_2$ ) at discharge, since this parameter is dominated by fission products. This indicates that no special handling, beyond what is currently practiced for discharged  $\text{UO}_2$  assemblies, is required for the discharged CORAIL assemblies. Since fuel handling is typically performed under water during refueling, the elevated neutron sources for the discharge state should not be a problem. In any event, it would be necessary to investigate spent fuel pool designs to confirm this conclusion.

At the reprocessing stage (5-year cooled fuel), the decay heat is highest when the minor actinides are recycled, again due to the high loading of Cm-244 and Pu-238. If aqueous processing is utilized, the high decay heat loads must be taken into account to ensure that organic solvents are not severely degraded. If centrifugal contactors are employed, as expected for advanced systems, the impact of the elevated decay heat level (a factor of ~3 over CORAIL-Pu) might be minimal, but must be evaluated.

The protection of workers at the reprocessing, fabrication, and reactor plants is most complicated by the high neutron emission rates which accompany minor actinide recycling. Multirecycling increases the Cf-252, Cm-244, and Cf-250 loading, and the corresponding neutron emission rates (and decay heat values), in the TRU fuel. Limiting the number of recycles might be necessary, but this will leave the TRU only partially consumed. Alternatively, lengthening the post-irradiation cooling interval to 20 years would allow the Cf-252 ( $T_{1/2} = 2.6$  years) and Cm-244 ( $T_{1/2} = 18$  years) to decay before reprocessing. A case assessing this assumption is also included in the results in Table 6.2. The neutron emission rate for the charge state of this case is a factor of 24 lower than that for the 5-year cooling case. Lastly, the curium could be removed from the CORAIL fuel cycle and stored, and only the neptunium and americium recycled with the plutonium. This would circumvent the buildup of Cm-244 and higher actinides in the CORAIL assembly, while still consuming the highly radiotoxic Am-241. Problems relating to criticality, heat load, and neutron emission rates would however have to be resolved for the curium storage facility in this approach.

It should be noted that the high heating rate and neutron emission rate provide intrinsic proliferation barriers. Table 6.3, shows that the gamma and neutron doses are much higher for this fuel compared to the CORAIL-Pu fuel. The spontaneous fission neutrons provide the major source of the dose rate (increasing from 0.014 to 1039 rem/hr). The contact doses for the CORAIL-TRU pellet and pin (about 1000-1500 rem/hr) are quite similar because the neutron dose is not significantly reduced by the presence of the zircaloy cladding as for the Pu-only case. Even at a distance of 1 meter away from the MOX pin, the dose is about ~5 rem/hr. While this is not self-protecting (defined as 100 rem/hr), it is quite higher than that for the Pu-only fuel, which is of the order of a few mrem/hr.

The removal of plutonium (except for separation losses) from the repository is also of non-proliferation advantage. Conversely, however, the application of TRU in the LWR fuel cycle will require additional fuels research and development efforts over the database existing in countries currently using MOX pins. This is because TRU-MOX pins have not been used for power generation in LWRs.

### **6.3 Pu+Np Separation**

The multirecycling of Pu+Np in the fuel cycle has been touted as an approach for decreasing the attractiveness of the material for weapons purposes and for providing

benefits to the repository by removing the Np-237 from the waste going to the repository. These issues are addressed in this sections.

The burning of Pu+Np in LWRs results in a significant reduction of the TRU mass to be disposed in a repository or to be burned in second-tier fast spectrum systems. Plutonium and neptunium constitute over 90% of the TRU content of spent nuclear fuel. The long-term radiotoxicity and decay heat arising from this concept are however quite similar to those of the Pu-only case (CORAIL-Pu). There are slight differences between the decay heat and radiotoxicity curves for the two cases, in the time frame after about 1,000,000 years (see Figs. 6.1 and 6.3), because of the elimination of the neptunium going to the repository (except from separation losses). The impact of removing neptunium from the spent fuel is however not pronounced. The long term radiotoxicity and decay heat contributions come primarily from the Np-237 produced from the  $\alpha$ -decay of Am-241. Therefore, as long as Am-241 is being buried in the repository, the long-term dose-rate hazards from the presence of Np-237, can only be marginally reduced.

The critical mass of a bare sphere of the TRU arising from plutonium and MA multirecycling has been found to be quite small (less than 20 kg); see Section 8.0. The case with Pu+Np multirecycling should result in a critical mass value between that of Pu-only and TRU recycling, thus indicating that the presence of Np in the fuel would provide no major impediment to the quantity of material required for a nuclear device.

The radiation source arising from the presence of neptunium is also not expected to be greatly increased. However, because neptunium in the fuel has been suggested as an approach for increasing the self-protection of the recycled LWR fuel (to elevate the radiation dose), this item has been separately evaluated in this work. The results of this study are provided in Appendix F. The expected increase in self-protection is attributed to the decay of Np-237 via  $\alpha$ -emission to protactinium-233 (Pa-233). The  $\beta$ -decay of the short-lived Pa-233 (half-life of 27 days) results in the emission of gammas (0.3 MeV), which are more energetic than those from reactor-grade plutonium decay chains. Therefore, the impact of the Pa-233 gammas depends highly on the level of shielding applied to the fuel.

The study found that mixing of the neptunium with the plutonium will not increase the contact gamma dose of the transuranic separation product, but will make the

gamma dose harder to shield; hence indicating that neptunium offers limited incremental protection to the fuel. The study reported above is for the initial state only.

Table 6.1 however shows that recycling neptunium in the fuel cycle increases the Pu-238 content in the charge state by about 70%, therefore, “denaturing” the plutonium vector. This increase in Pu-238 translates to elevated levels of the decay heat and gamma sources for the Pu+Np recycling case (see Table 6.2). Using the gamma dose rates in Appendix G and the vectors in Table 6.1, it was determined that the gamma dose would be increased by about 70%, and the neutron emission rate by ~50% at the charge state, compared to the CORAIL-Pu case. The results in Table 6.3, which are for detailed estimation of the radiation dose rates arising from contacts at pellet and cladding surfaces, confirm these estimated values. The gamma dose dominates in this fuel form (>99% at the pellet surface and 65% at the pin surface). Shielding with 0.06 cm of zircaloy cladding helps to reduce the pin surface dose to below 1 rem/hr (from > 5 rem/hr for the pellet). With this material however, the relative contribution of the neutron dose is increased, presumably as a result of the increased neutron multiplication of the pin versus pellet and the difficulty of shielding neutrons. At 1 meters away from the pellet and pin, the dose rate is much diminished (< 1mrem/hr).

Taken together the dose rate estimations indicate an elevation of the fuel dose rate with Pu+Np recycling, arising mostly from the denaturing of the Pu-vector, not the presence of Pa-233 produced from the decay of Np-237. The data shows that the fuel pellet and pin are however not self-protecting, as the dominant radiation sources (fission products and higher actinides) have been separated.

#### **6.4 Pu+Np+Am Separation**

Multirecycling of the plutonium, neptunium and americium contained in spent nuclear fuel provides benefits to the repository, beyond those indicated for Pu-only recycling, as would be expected. In particular, the minimization of the americium going to the repository removes Am-241, which is the major intermediate term contributor to the radiotoxicity (see Figure H.1 in Appendix H for UO<sub>2</sub> fuel). Additionally, Am-241 is the primary source of Np-237, which is a major contributor to the long-term radiotoxicity and dose rate outside the repository setting, as a result of its high mobility. Fig. 6.1 indicates that for the Pu+Np+Am scenario, the long-term radiotoxicity in the repository is significantly reduced compared to the base UO<sub>2</sub> case. Fig. 6.3 shows that the long-term decay heat is also quite reduced compared to direct disposal in spent nuclear fuel (UO<sub>2</sub> case).

The repository benefits derived from the removal of americium is however diminished by the presence of curium in the fuel. Curium-244 ( $T_{1/2} = 18$  years) results in the creation of Pu-240 ( $T_{1/2} = 6,560$  years), and prolongs the time required to reach the source uranium radiotoxicity level. This trend is evident in Fig. 6.2, which displays the contributions of the leading isotopes to the radiotoxicity.

The impacts of Pu+Np+Am multirecycling on the fuel cycle are shown in Table 6.2. At the fabrication and charge states, the decay heat level is elevated by a factor of ~6, relative to the CORAIL-Pu case. The relative increase is about 2 in the separation stage (i.e., 5 years after cooling). The gamma source level at the fabrication and charge stages are also relatively increased by factors of 13.7 and 8.5, respectively. Estimations using the data in Appendix G, indicate that the contact gamma dose rate would be increased by a factor of about 10 in the charge stage, relative to the CORAIL-Pu case. These elevations in the gamma source and dose, over that of the Pu-only and Pu+Np fuel forms arise because of the presence of Am-243 and Am-241. The combined dose contribution from the americium isotopes is greater than that from Pu-238, in the Pu+Np+Am fuel form.

Neutron emission rate (and hence dose) are also increased in the fuel cycle by factors between 2 and 5. In the Pu+Np+Am fuel form however, the plutonium isotopes are the leading sources of neutrons, since Cm-244 and higher isotopes are not recycled. The gammas are the leading contributors to dose from exposure to the fuel.

The equilibrium recycling of Pu+Np+Am results in significant increase of both the uranium enrichment in the  $\text{UO}_2$  pins and in the TRU content of the MOX pins (Table 6.1). These trends indicate slight degradation of the assembly reactivity coefficients, but these differences are not expected to adversely affect core safety. The increased enrichment (4.9 to 5.5%) and MOX content (10 to 16%) are expected to increase fuel assembly costs, relative to the Pu-only recycling case. Additionally, significant fuel development activities are expected for the application of the Pu+Np+Am fuel form.

This separation hypothesis (Pu+Np+Am) is not without practical problems. Because americium and curium both exist in the trivalent state, there are difficulties in separating them from each other. A separation process for these elements has been conceived in France, but cost and complexity make it currently unattractive.

### **6.5 Np and Fractional Pu +Am Separation**

Figures 6.2 and 6.4 show that Pu-240 dictates the intermediate term (300 to 10000 years time scale) radiotoxicity and decay heat in the repository, for the case with

Pu+Np+Am recycling. The Pu-240 arises from the decay of Cm-244. So the next logical question is the feasibility of minimizing the curium going to the repository in order to provide some benefits to the repository. (This is clearly possible if curium is additionally burned in the fuel cycle, however this results in the elevation of radiation dose, which might be problematic to fuels workers). It has been suggested that the removal of Pu-242 or both Pu-242 and Am-243 (to minimize Cm-244) in the fuel cycle by isotopic separation, might be an option. An evaluation of these options (no Pu-242 and no Pu-242+Am-243 in the Pu+Np+Am recycle scenario) has been done.

The results for the fractional separation cases are included in Tables 6.1 and 6.2, and Figs. 6.1 and 6.3. The Figures indicate that the fractional separation of plutonium and americium provides intermediate term benefits to the repository. However, it is noted that isotopic separation on the required scale is a major technological challenge.

Pu-242 and Am-243 constitute 19% and 5% of the charge Pu+Np+Am fuel from discussed in Section 6.4. Removing these nuclides results in a relative increase in the fraction of the other transuranics. However, because Pu-242 is a strong absorber in the thermal spectrum, its removal and the relative increase in the fissile content, results in a decrease in the TRU content of the MOX fuel. This decreases TRU separation costs; isotopic separation would however increase cost, and must be factored into the overall cost. Fuel development research is also required for this fuel form.

The removal of Pu-242 and Am-243 was found to decrease the radiation sources and doses in the fuel cycle, relative to the case for the Pu+Np+Am fuel. The removal of Am-243 is beneficial to the reduction of the gamma dose (about half). The other fuel handling indices and gamma dose rate are reduced (relative to the Pu+Np+Am fuel) because of the lower TRU content in the MOX pins (9-10% versus 16%).

While the partial separation of plutonium and americium offers benefits to the fuel cycle and the repository, there are issues that have to be addressed. First, isotopic separation on the scale of interest would be a major undertaking. Additionally, as for the Pu+Np+Am case, the separation of americium from curium is a costly endeavor that requires advances in separation technology for the process to be viable. Also, development of the waste forms for the separated nuclides and the future storage of these materials have to be addressed.

## **6.6 Lessons Learned for the Separation Hypotheses Study**

The results presented here contain enough data that could be used to draw conclusions for the best approach for LWR-based transmutation of waste. The approach

must resolve the conflict between recycling americium and curium in the fuel cycle (provide measurable benefit to the repository) and that of not recycling curium to provide benefit to the fuel cycle campaigns (i.e., minimize fuel handling problems).

The recycling of neptunium in the fuel cycles provides incremental benefit to the proliferation resistance of the fuel cycle, because of the denaturing of the plutonium vectors (relative increase in Pu-238). However, the dose rates do not increase more than a factor of two over that of the Pu-only recycling case and both of these fuel forms are not self-protecting. The additional recycling of neptunium, while providing additional mass reduction, has minimal effect on the radiotoxicity, dose rate, and long-term heating rate, compared to the Pu-only case. So this option provides only marginal benefits.

Additionally recycling Am with Pu+Np provides significant benefits to the fuel repository, the fuel cycle, and the proliferation resistance of the fuel. Compared to fuel cycle results for the Pu-only case, the decay heat levels are increased a factor of 2 to 6, the neutron source/dose by a factor of 2 to 4, and the gamma dose by a factor of ~10 at the fabrication and charge states. The benefit provided by this approach is however limited if Cm-244 is buried in the repository, because of the decay of this nuclide to Pu-240, which dictates the intermediate term radiotoxicity and heat loads. The partial separation of plutonium and americium by isotopic separation, would help in this regard, as the amount of Cm-244 to be passed to the repository would be decreased. The fuel handling indices (radiation sources and decay heat) are however reduced, primarily because the absence of Pu-242 reduces the TRU content of the fabricated MOX pin. There are however technological challenges associated with the separation of americium from curium that have to be resolved. Additionally, the cost of isotopic separation and of storing the curium and Pu-242 and/or Am-243 could become unattractive and make these approaches untenable.

TRU recycling provides the best benefit to the repository, both in terms of the radiotoxicity and dose rate, and the heat load. While this option is also beneficial from proliferation resistance viewpoint, its application would complicate fuel handling, and at the least make it most expensive, because of the shielding requirements, in particular in the fuel fabrication plant. It would be useful to have a comparative analysis of this shielding cost to that of multirecycling of TRU in a fast reactor fuel cycle. It is anticipated that remote fuel handling would be required in any event for TRU multirecycling in LWR systems.

In regards to fuel handling issues, the preferred approach is to recycle plutonium, neptunium, and americium, and then store curium (taken as Cm-244 in this discussion) for a few of its half-lives. This provides both proliferation resistance and repository benefits. With this approach the radiotoxicity curve would be similar to that for the CORAIL-TRU case. Table 6.4 shows that if Cm-244 is stored for 360 years, which would be in the time scale of the period in which fission products dominate the decay curve, nearly all the Cm-244 would have decayed to Pu-240. Following this time, the Pu-240 could be burned as special targets in the nuclear fuel cycle. The modalities for Cm-244 storage outside the repository would have to be evaluated. However, because of its high specific neutron source value, such a storage facility will require sufficient shielding and result in added cost to the fuel cycle. Also, with curium (not only Cm-244), criticality issues have to be addressed, along with the problem of heat removal in the storage facility.

In closing, another proposed alternative is for the curium and americium to be burned as separated target pins (i.e., heterogeneous recycling) in nuclear systems [14]. This has the potential of confining the problems associated with their use in reactor systems and the fuel cycle. In this regard, neutron-inert materials like MgAlO<sub>3</sub> (spinel) have been suggested as a medium for burning the material. However, heterogeneous recycling is limited by the fission rate to be attained. If the fission rate is ~90% (already very high, and with attendant high damage rates, since the irradiation needed is 20 years), the radiotoxicity is reduced only a factor of 20-30 with respect to the open fuel cycle.

**Table 6.1. Comparison of Equilibrium State TRU Content of Initially Charged CORAIL Fuel.**

Target TRU			Pu	Pu-Np	Pu-Np-Am	No Pu-242 <sup>a)</sup>	No P2/A3 <sup>b)</sup>	TRU	TRU
Cooling time, year <sup>c)</sup>			5	5	5	5	5	5	20
U Enrichment, %			4.62	4.9	5.5	5.5	5.5	5.12	5.12
TRU content, %			8.45	9.93	16.17	9.77	9.26	20.39	20.39
Fissile, %			44.67	43.22	36.23	49.23	50.39	32.80	28.87
Np237			1.66E-3	2.84	2.23	3.02	3.12	1.82	2.39
Np239			0.00	0.00	0.00	0.00	0.00	0.00	0.00
Pu238			3.55	6.13	10.95	13.40	13.60	10.20	11.11
Pu239			34.20	33.25	28.27	38.09	39.33	24.40	24.80
Pu240			23.33	22.38	20.55	25.12	25.16	21.67	23.22
Pu241			10.48	9.98	7.89	11.08	11.43	7.13	3.24
Pu242			27.40	24.44	18.99	0.00	0.00	16.93	16.34
Am241			1.04	0.99	6.01	7.22	7.29	5.91	10.34
Am242m					0.06	0.06	0.06	0.07	0.10
Am243					5.05	2.00	0.00	4.59	4.49
Cm242								0.00	0.00
Cm243								0.03	0.03
Cm244								3.90	1.51
Cm245								1.14	0.68
Cm246								1.74	1.33
Cm247								0.15	0.12
Cm248								0.29	0.27
Bk249								3.00E-5	0.0E+0
Cf249								1.91E-2	1.91E-2
Cf250								3.48E-3	1.57E-3
Cf251								4.95E-3	5.35E-3
Cf252								9.40E-4	3.00E-5
Cf253								0.0E+0	0.0E+0
Es253								0.0E+0	0.0E+0
Mass Balance (kg per assembly)	Target TRU	Charge	14.7	17.2	28.3	17.1	16.1	35.7	35.7
		Discharge	15.2	17.8	28.2	17.1	16.1	36.0	36.5
		Net	0.5	0.6	-0.1	0.0	0.0	0.3	0.8
	Other TRU	Charge	0.2	0.2	0.0	0.0	0.0	0	0
		Discharge	1.5	1.3	1.1	1.3	1.4	0	0
		Net	1.3	1.1	1.1	1.3	1.4	0	0
	Total TRU	Charge	14.9	17.4	28.3	17.1	16.1	35.7	35.7
		Discharge	16.7	19.1	29.3	18.4	17.5	36.0	36.5
		Net	1.8	1.7	1.0	1.3	1.4	0.3	0.8

a) Multi-recycling of Pu, Np and Am without Pu-242.

b) Multi-recycling of Pu, Np and Am without Pu-242 and Am-243.

c) The cooling time between assembly discharge and fuel separation.

**Table 6.2. Comparison of Equilibrium State Radiation Parameters for Fuel Handling Assessment.**

	Assembly	CORAIL-Pu	CORAIL-TRU		Pu-Np	Pu-Np-Am	No-Pu242 <sup>a)</sup>	No-Pu2/Am3 <sup>b)</sup>
	Cooling time (yr)	5	5	20	5	5	5	5
Decay Heat (Watt)	Fabrication	1960	38390	25520	3766	11600	8531	8138
	Charge	655	11930	8278	1249	3788	2791	2662
	Discharge	2002000	1999000	2013000	2008000	1993000	1994000	2000000
	After cooling	4404	14520	9118	4924	9551	5925	5119
Neutrons (n/sec)	Fabrication	1.20E+08	7.65E+12	2.45E+11	1.84E+08	4.60E+08	3.01E+08	2.87E+08
	Charge	3.96E+07	1.51E+12	6.22E+10	6.05E+07	1.50E+08	9.82E+07	9.36E+07
	Discharge	9.59E+09	9.08E+12	1.25E+13	9.45E+09	2.50E+10	1.08E+10	7.26E+09
	After cooling	6.36E+09	2.51E+12	1.07E+11	6.00E+09	1.51E+10	4.47E+09	1.70E+09
Gamma Source (Watt)	Fabrication	0.57	23.30	20.67	1.08	7.79	5.36	4.98
	Charge	0.32	7.10	9.13	0.51	2.73	1.91	1.79
	Discharge	547725	533898	533738	549022	538192	542213	546283
	After cooling	1030.14	978.56	344.39	1020.30	982.25	1010.77	1009.67

a) Multi-recycling of Pu, Np and Am without Pu-242.

b) Multi-recycling of Pu, Np and Am without Pu-242 and Am-243.

Note:

1. The TRU vector used for estimating these parameters were obtained from TRANSEQM calculations.
2. The fabrication stage data is for one metric ton of MOX pin. Charge data is based on one metric ton of heavy-metal (U and TRU) present in the charge assembly. Data for discharge and after cooling (five or 20 years) are based on the resulting mass after irradiation (~0.954 ton in all cases). The discharge and five (or 20) year after discharge (after cooling) data include contributions from fission products.
3. Gamma source is expressed in equivalent MeV/s (or watts), as a means of providing indications of the relative unshielded gamma dose rates. This approach does not capture the overall gamma dose, however, particularly for shielded situations.

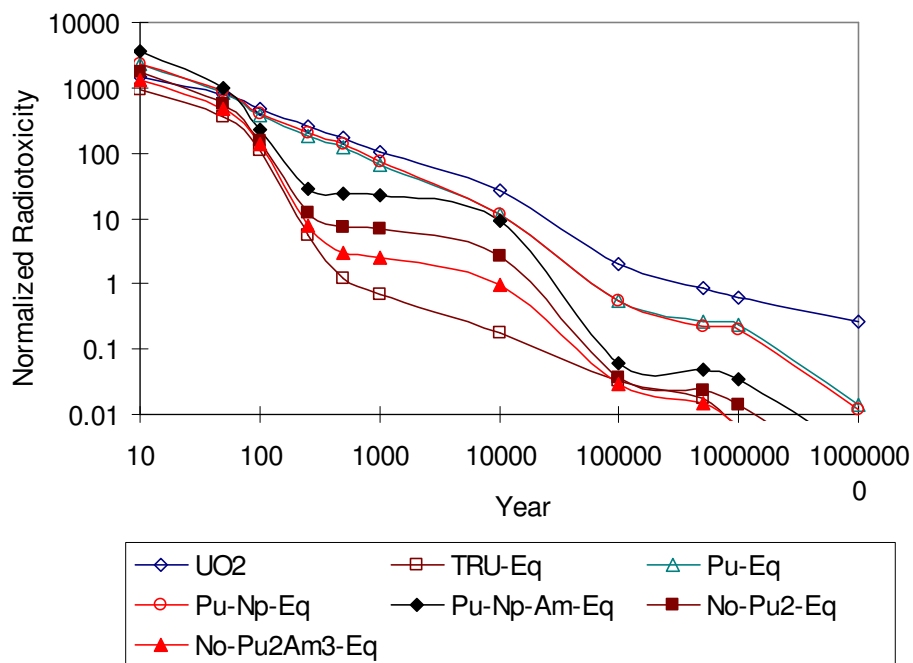
**Table 6.3. Comparison of Dose Rates of Equilibrium State CORAIL MOX Fuels at Charge Stage (rem/hr).**

		CORAIL-Pu	CORAIL Pu+Np	CORAIL-TRU
Surface of pellet	Spontaneous Fission	$0.014 \pm 0.001$	$0.017 \pm 0.001$	$1039 \pm 0.001$
	( $\alpha$ ,n)	$0.016 \pm 0.001$	$0.032 \pm 0.001$	$0.34 \pm 0.001$
	Photon	$4.893 \pm 0.019$	$8.776 \pm 0.020$	$110.6 \pm 0.016$
	Total	4.9	8.8	1149.9
Surface of fuel cladding	Spontaneous Fission	$0.019 \pm 0.003$	$0.024 \pm 0.003$	$1436 \pm 0.003$
	( $\alpha$ ,n)	$0.021 \pm 0.003$	$0.041 \pm 0.003$	$0.43 \pm 0.003$
	Photon	$0.094 \pm 0.134$	$0.124 \pm 0.176$	$18.46 \pm 0.040$
	Total	0.13	0.19	1454.9
One meter away from the surface of cladding	Spontaneous Fission	$6.13\text{E-}05 \pm 0.003$	$7.56\text{E-}05 \pm 0.003$	$4.61 \pm 0.002$
	( $\alpha$ ,n)	$6.22\text{E-}05 \pm 0.003$	$1.23\text{E-}04 \pm 0.003$	$1.29\text{E-}03 \pm 0.003$
	Photon	$2.96\text{E-}04 \pm 0.099$	$3.28\text{E-}04 \pm 0.129$	$0.06 \pm 0.037$
	Total	$4.2\text{E-}04$	$5.3\text{E-}04$	4.7

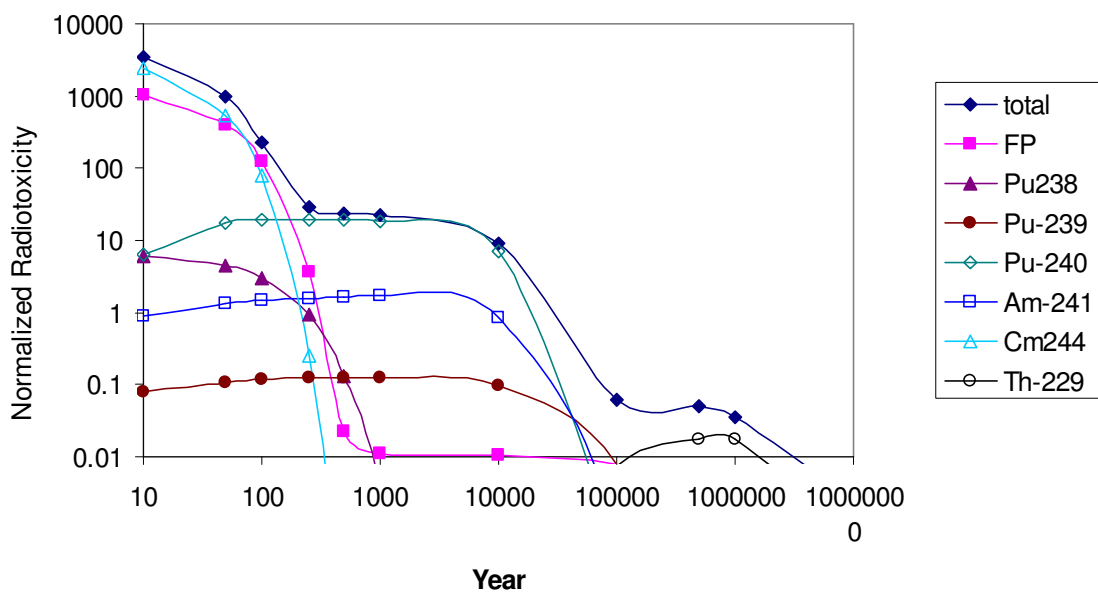
a) Heavy metal isotope vector was calculated by the TRANSEQM code.

Note:

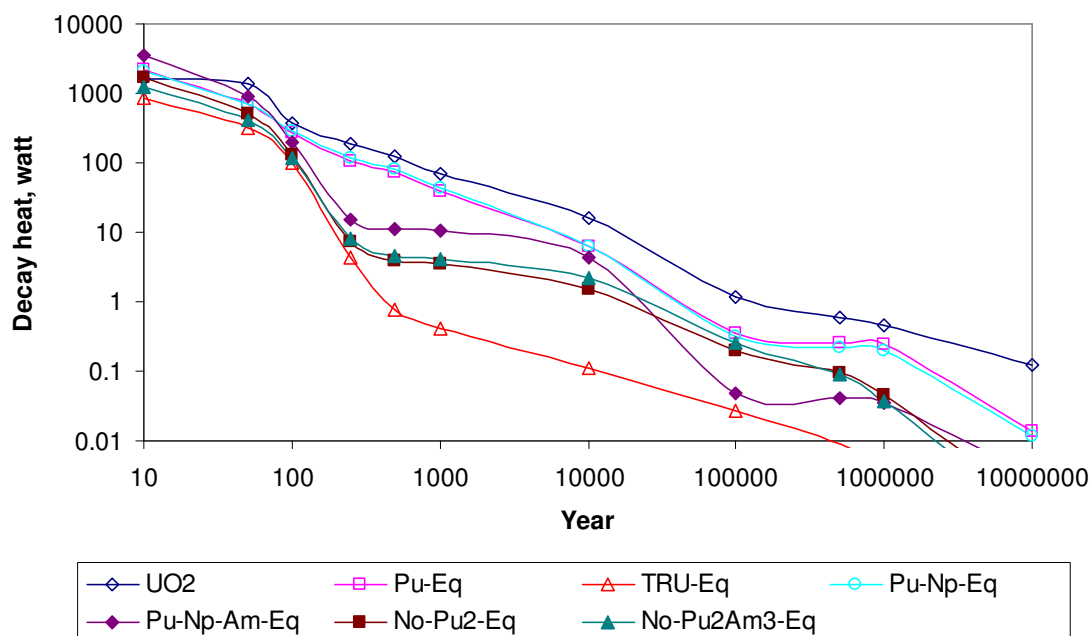
1. Flux-to-dose rate conversion factors for neutrons and photons are from ANSI/ANS-6.1.1-1977 (See Ref. 4).
2. Spectrum for the spontaneous fission neutrons is from MCNP4 and that for photons comes from ORIGEN2 calculations.
3. Spectrum for ( $\alpha$ ,n) neutrons is based on generic data for MOX fuel (see Appendix E).
4. Number of neutron histories =  $1.0\text{E}+06$ ; photon =  $5.0\text{E}+06$ .



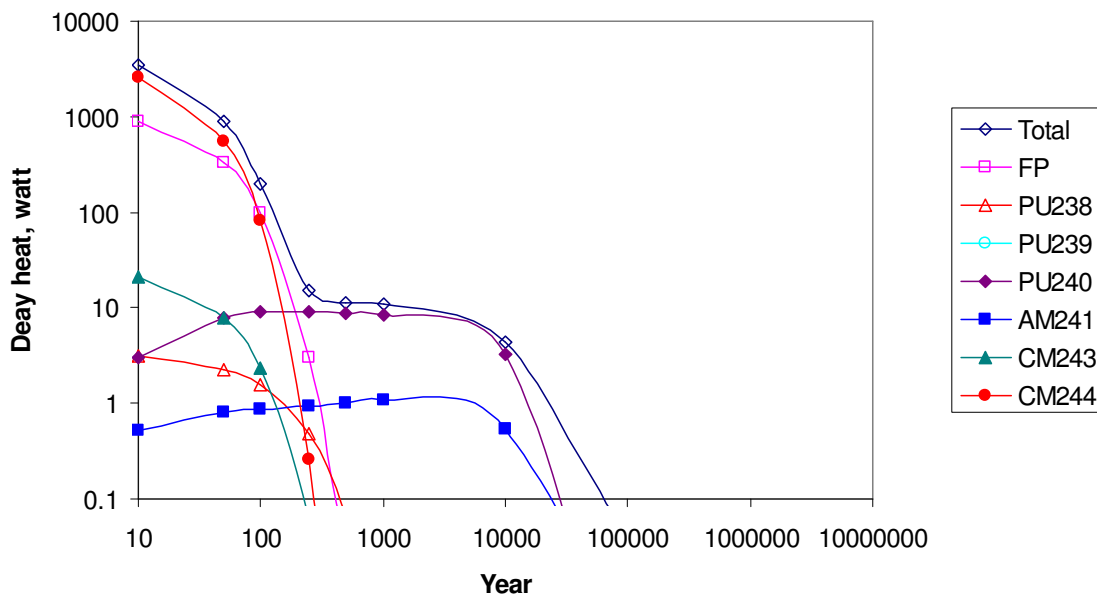
**Fig. 6.1. Comparison of Normalized Radiotoxicity at the Repository.**



**Fig. 6.2. Comparison of Normalized Isotopic Radiotoxicity at the Repository from Pu+Np+Am Recycling.**



**Fig. 6.3. Comparison of Decay Heat at the Repository (per One Ton).**



**Fig. 6.4. Comparison of Isotopic Decay Heat at the Repository from Pu-Np-Am Recycling (per One Ton).**

**Table 6.4. Impact of Decay Time on Cm-244 Content.**

Number of Half-Lives	Time Elapsed (Years)	Relative Value
0	0	1.0000
5	91	3.E-02
10	181	1.E-03
20	362	1.E-06

## 7.0 Practical Number of Stages for CORAIL-TRU Multirecycling

The results presented in Section 6.0 and Ref. 3 indicate that from a core performance (reactivity balance and safety coefficients) point of view, the transmutation goal of attaining a radiotoxicity level comparable to that in the source uranium can be satisfied by using the CORAIL assembly for TRU multirecycling. Additionally, with this approach, the long-term dose rate (absence of Np-237) and heat loads are also reduced. However, there are issues relating to the elevated minor actinide content with recycling and its attendant fuel handling issues, that might limit the number of recycle stages.

Equally important to the overall transmutation strategy is the time required to complete each CORAIL-TRU recycle campaign – roughly 4.5-year core residence, 5-year cooling, and 2-year separation/fabrication. During this time the TRU stockpile inventory is roughly conserved. Therefore, a limited CORAIL recycle could be employed to mitigate the short-term growth of TRU/Pu inventories; this delay allows time to deploy an advanced fuel cycle system (including a fast-spectrum transmuter) for extended recycle and waste transmutation. In this Section, an attempt is made to quantify the number of practical recycle stages that would minimize the burden on the fuel cycle. This is done by evaluating cycle by cycle data for the CORAIL-TRU assembly. The first seven recycle stages have been evaluated and compared to the results for CORAIL-Pu, which appears feasible compared to a full MOX loading core. [2]

The mass values for each recycle stage were calculated using a coupled WIMS8-TRANSEQM procedure. In this procedure, both codes are utilized to predict the isotopic masses in the fuel assembly. However, because the transmutation chain of the WIMS8 code ends at Cm-245, the isotopic masses tracked by WIMS8 code are re-normalized by the total mass of these isotopes predicted by the TRANSEQM code. The masses that TRANSEQM calculated for nuclides above Cm-245 are used without modification. In this coupling procedure, the cross sections for the TRANSEQM calculations were obtained from WIMS8 calculations. The results from this coupling procedure are combined with those from ORIGEN2 to obtain consistent data for the decay heat and radiation sources. MCNP4 calculations are additionally performed, using radiation source terms from ORIGEN2, to obtain the dose rates at the surfaces of the fuel pellet and pin.

Table 7.1 is a summary of the uranium enrichment, plutonium content, fissile content, and heavy-metal vector for the beginning of cycles 2 to 7, predicted by the WIMS8-TRANSEQM procedure; the CORAIL-TRU masses tracked by the TRANSEQM code only, are provided in Appendix K. The cycle 1 data (not shown) is

that of the original vector derived from the spent nuclear fuel (SNF) of currently operating reactors. Data for cycle 2 represent the vector recovered from one recycle of the original spent nuclear fuel (SNF) in a CORAIL-TRU assembly. This vector accounts for a five-year cooling period, a 0.1% loss of TRU or plutonium in the separation stage, and an additional two-year period following fuels separation and fabrication.

The data in Table 7.1 indicate that the fissile content of the CORAIL MOX pins decreases with recycle stage as expected, since the fissile component of the fuel is burned preferentially in the thermal spectrum of the reactor. Because of the decrease in the fissile content, the plutonium or TRU contents of the MOX pins also increases with recycle stage, and it is about 12w/o in recycle stage 7 of the CORAIL-TRU approach.

It should be additionally noted that the charged fraction of the higher actinides (above Cm-245) increases with recycle stage. This component has been neglected in previous results reported for the CORAIL-TRU assembly [3,15]. The main impact of this improved modeling is the increase in the spontaneous neutron source arising from contributions from Cf-252 (primarily), Cf-250 and Cm-246. In fact, Cf-252 is the dominant contributor to the neutron source in the equilibrium state of the CORAIL-TRU multirecycling concept. As can be observed in Table 7.1, the mass fraction of this nuclide is quite small, but compared to Cm-244, its specific neutron source (neutrons/kg-s) is a factor of 215,000 times that of Cm-244. As a result extreme care was taken to ensure that numerical round-off error does not overwhelm the Cf-252 mass value predicted by the TRANSEQM code. The impact of the assumed cross sections on this value is an item that requires additional investigation.

The decay heat, neutron sources, and gamma source at difference stages of the fuel cycle have been estimated for the CORAIL Pu and TRU assemblies. These data have been compared to those for the 4.0w/o enriched UO<sub>2</sub> fuel assembly and for a traditional MOX assembly containing 9.4w/o plutonium derived from LWR SNF. The results of these comparisons are provided in the following sub-sections. The results are based on one ton of the heavy metal in the fuel material, pin or assembly, depending on the point in the fuel cycle. For the discharge fuel, the mass is that resulting from the fuel burnup (~0.954 tons; 45 GWd/t burnup). Additionally, the case for recycle stage 7 of the CORAIL-Pu assembly has been taken as reference for comparison (similar to the equilibrium state in this case). Note that one ton of heavy metal corresponds to about the total heavy-meal mass contained in two assemblies (535 kg each).

## 7.1 Comparison of Decay Heat Values

Table 7.2 provides a comparison of the decay heat values for the different cases and stages. It was found that the leading contributors in charged plutonium fuel (i.e., MOX and CORAIL-Pu assemblies) are Pu-238 (80%), Pu-240 (10%), and Pu-239 and Am-241 (about 5% each); see Appendix D for nuclides. For the charged TRU fuel, the leading contributors to the decay heat are Cm-244 (54-64%) and Pu-238 (about 30%).

For the charged CORAIL-Pu assembly note that only 30% of the fuel pins are MOX pins, while it is 100% in the MOX assembly case. This is the reason for the factor of three difference in the decay heat of the MOX assembly relative to the CORAIL-Pu case for the fresh (charge) assembly. The large difference in the decay heat levels of fresh fuel in the CORAIL-Pu and TRU assemblies is due to the absence of Cm-244 in the CORAIL-Pu assembly. The value of the decay heat increases with the recycle stage and in cycle 7, the CORAIL-TRU assembly has a value that is a factor of 10 greater than that of the CORAIL-Pu fuel that has been recycled for the same amount of passes through the reactor (see Fig. 7.1).

In the 5-year-cooled MOX, CORAIL-Pu and CORAIL-TRU fuel assemblies, Cm-244 is the leading contributor with Pu-238, Cs-134, Ba-137m (from Cs-137), and Y-90 (Sr-90). For the UOX assembly however, the fission products dominate.

The results show that the decay heat values for the MOX and CORAIL assembly are quite larger than that for the UO<sub>2</sub> assembly. This implies that additional considerations be given to fuel cycles using recycled plutonium, particularly in the fuel separation stages, where increased heating might have effects on the organic solvents employed. Previous analysis suggests that with the use of modern equipment such as centrifugal contactors and pin choppers, this might not be a problem; with the modern contactors, the spent fuel and the solvent would be in contact for a short time. [15] If necessary, the CORAIL-TRU fuel assembly may be cooled longer to ensure that the decay heat is sufficiently reduced prior to reprocessing. Additional review of open literature revealed that the decay heat levels obtained at the reprocessing stage (after five-year cooling) for CORAIL-TRU cycle 7 (10 W/kg-HM) is quite lower than those for plutonium burner fast reactor (21.8 W/kg-HM) and TRU burner fast reactor (46.0 W/kg-HM). The decay heat levels in these fast spectrum designs were not considered to be problematic in the OECD/NEA study (page 62 of Ref. 14).

Heat removal during fuel fabrication is an issue that has to be given additional consideration. The data in Table 7.2 indicate that, for the TRU fuel material, decay heat

by the seventh cycle will be ten times greater than that for a traditional MOX assembly, due to the presence of Cm-244 in the TRU assembly. Analysis of fuel fabrication components would be required to determine the temperatures to be expected from this increased heat source in the fuel material. If any component temperature limits are predicted to be exceeded, means of cooling these components will have to be developed if the hotter fuel material is to be used to fabricate new pins. [15]

## 7.2 Comparison of Neutron Sources

Results for the combined ( $\alpha$ ,n) and spontaneous neutron sources are compared in Table 7.2 for the different recycle stages and cases. Analysis revealed that the leading contributors in the charged plutonium fuel are Pu-238, Pu-240, and Pu-242. In the CORAIL-TRU fuel, it is predominantly Cm-244 (85%) for the early recycle stages and Cf-252 and Cm-244 (combine > 90%) from stages 5 to 7. These isotopes are also the primary contributors to the neutron source of the 5-year cooled fuel.

When neutron sources for the charged MOX and CORAIL-Pu assemblies are compared, the expected factor of three is actually 2.7 because of the higher Pu-242 content of the CORAIL-Pu assembly.

Similarly to the decay heat results, the CORAIL-TRU assembly has a much higher neutron source level than the CORAIL-Pu and MOX assemblies, especially for the charged fuel. This is because of the presence of Cm-244 and Cf-252 in the CORAIL-TRU fuel. The neutron source of the charged CORAIL-TRU assembly is a factor of ~80 to 1540 (in cycle 7) higher than that of the CORAIL-Pu assembly. The differences between the cases are lower (factors of 1.5 to 50) for the discharge and 5-year cooled stages because of the presence of the minor actinides at these stages, for all the cases.

The implication of the high neutron source levels in the CORAIL-TRU fuel is addressed in Section 7.4.

## 7.3 Comparison of Gamma Sources

Gamma source levels are compared in Table 7.2 for all the cases and stages. The gamma source is displayed in the unit of Watts (equivalent MeV/s), because this unit provides a better indication of the gamma dose to workers, particularly for the unshielded fuel; it does not however capture the overall effect. The leading contributors to the gamma source of the charged plutonium fuel are Pu-238 (50%) and Am-241 (40%). For the charged TRU fuel, they are Am-241 (30-70%), Cm-244 (~20-30%), and Pu-238 and Cm-243 (9-18% each).

In the 5-year cooled fuel, Cs-134 and Ba-137m account for about 80% of the gamma source. At this state, fission products dominate and hence the similarity in the values for all the cases, including the UO<sub>2</sub> assembly. The value for the UO<sub>2</sub> fuel is slightly higher because the Cs-134 and Ba-137m fission yields are higher for uranium fuel than for TRU fuel. The ORIGEN2 calculation indicated that the mean gamma energy is 0.014 to 0.02 MeV for the fresh MOX and CORAIL assemblies, 0.09 MeV for the fresh UO<sub>2</sub> assembly, and between 0.3 and 0.4 MeV for 5-year cooled fuel. The different values are indicative of the different gamma energies accompanying the decay processes. The leading fission products in the five-year cooled fuel give relatively high energy gammas. The implications of the gamma levels are discussed in the next section.

#### **7.4 Evaluation of Radiation Doses for Fuel Pellet and Pin**

Radiation doses arising from gamma and neutron sources presented in Sections 7.2 and 7.3 have been evaluated for fuel pellet and pin configurations. These different configurations are used because it is expected that cladding material would provide shielding that is not available for a pellet. The bulk of the assembly would give additional shielding for the gammas, but could increase the dose for the neutrons because they are harder to shield and because of increase in the subcritical multiplication relative to the pin or pellet.

MCNP4 models for the different geometry were created and used to track the gamma and neutron source particles as they interact with the material and external medium (taken as air). The fuel material is assumed to be in oxide form, and so both spontaneous fission and neutrons from ( $\alpha$ ,n) interactions were considered, in addition to the gammas. Calculations were done for the second and seventh recycle stages and the equilibrium state of the CORAIL-TRU, and for the first, seventh, and equilibrium states of the CORAIL-Pu case. Results for these cases are summarized in Table 7.3.

Dose rate calculations for all the stages have not been done in this study because the dose rates correlate with the gamma and neutron sources. For example the ratios of the pellet gamma dose rates for CORAIL-Pu and CORAIL-TRU cycle 7 are 10.3 from Table 7.2 (using the gamma source) and are 10.7 from Table 7.3 results. Similar ratios for the neutron dose rates are 2185 from Table 7.2 (using neutron source) and 1232 from Table 7.3. The factor of two discrepancy in the neutron dose rates is due to the fact that the ( $\alpha$ ,n) and spontaneous fission source levels are similar, whereas the spontaneous fission dominate the dose rate. This is because of the higher average neutron energy for

the latter (2 MeV for spontaneous fission neutrons and 0.1 MeV for neutrons from the  $\alpha, n$  interaction in the oxide fuel [16]).

The results in Table 7.3 indicate that the values obtained for the seventh and equilibrium stages of the CORAIL-Pu are quite similar, justifying the use of cycle 7 for approximating the equilibrium state, in the previous Sections (7.1 to 7.3). Additionally, it is noted that the dose rates for the CORAIL-Pu MOX fuel is dominated by the gammas (photons). In fact, for the CORAIL-Pu fuel the gammas contribute nearly all the pellet-surface dose (>99%).

At the equilibrium state of CORAIL-TRU, neutrons dominate the dose rate. Up to seven recycle stages, the gamma dose is the largest contributor to the overall pellet-surface dose for the CORAIL-TRU fuel. The magnitude of the gamma dose is greatly affected by the presence of a 0.06 cm thick zircaloy cladding, which provides some shielding against the gammas. The cladding has little impact on the neutron dose. Therefore for the pin, the ratio of the neutron and gamma doses is about 0.4 for the CORAIL-Pu stages, and ranges from 1.7 to 7.5 for the CORAIL-TRU stages. In the CORAIL-TRU equilibrium state, the spontaneous neutron source accounts for 99% of neutron dose on the pin surface. At a distance of 1 meter away from the pin surface, the dose rates for the CORAIL-Pu and TRU assemblies are relatively small (about 0.4 mrem/hr and 200 mrem/hr, respectively, for stage 7). The equilibrium state of the CORAIL-TRU gives a value of about 4,700 mrem/hr.

The results presented here indicate that using moderate thickness of shielding material (e.g. 0.5 to 1.0 cm thick lead shield) the gamma dose could be significantly reduced. Similarly, using appropriate shielding for neutron during fabrication and fuel loading at the plant should make it possible to achieve seven recycles of the TRU in the CORAIL assembly. The shielding material could include a combination of neutron moderator and absorber.

It should be noted that the neutron emission rate of the fresh assembly in CORAIL-TRU stage 3 (Table 7.2) is comparable to that of the 5-year cooled spent fuel assembly of the full-MOX assembly. Since this latter fuel has to be handled for reprocessing or dry storage in the traditionally MOX fuel cycle, this might provide an indication of the number of recycle stages that is reasonable in the CORAIL-TRU concept. Note however, that even for 3 recycle stages, the shielding requirements would make fuel handling more cumbersome and costly. Additionally, because of the elevated neutron source in the 5-year cooled fuel after 3 recycle stages, the redesign of the spent

fuel pond might also be necessary if the original design was optimized to accommodate the neutron dose load expected from  $\text{UO}_2$  fuel assemblies.

Three recycle stages in the current CORAIL-TRU concept requires about 35 years to complete. The actual amount of time would be longer since the starting time for the implementation of the concept over the U.S. commercial nuclear complex would take longer, as plants have to be staggered in time to ensure that fuel is available for the next stages. The results presented in Section 6.0 suggest that if the cooling time is increased to 20 years (from 5 years), charge neutron source can be reduced by a factor of 24 (in the equilibrium state). Therefore, providing the opportunity for more recycle stages and a longer time for development of advanced transmutation systems that can perform the mission more efficiently and economically.

The utilization of limited recycling (e.g., 3 stages) of legacy TRU in the CORAIL assembly has the advantage that the fuel would be used to generate electricity. However, since the CORAIL concept is a delay line, the overall mass of the legacy waste would not be decreased and more fission products would have been produced. Additionally, the minor actinide content of the fuel would be higher, therefore, making it more radiotoxic than the original legacy TRU. Therefore, the most likely solution for this spent fuel is to burn it in a fast reactor system, in order to complete the transmutation mission. An analysis of this case is currently ongoing under the AAA Downselection Studies. Preliminary results suggest that the multirecycling of the recovered TRU in the fast reactor is feasible. However, material handling and processing would have to be done remotely. It has been proposed that after the three recycles, the fuel could be stored for a longer cooling time to reduce the heat and radiation emission loads on the fast reactor fuel cycle. This long cooling time is an advantage, as it allows time for the deployment of fast reactors or other advanced systems.

It is noted that the neutron source levels at the fabrication stage (10 to 260 n/kg-s HM) for CORAIL-TRU is lower than those reported for the TRU burner accelerator driven system (670 n/kg-s HM) and minor actinide burner ADS (1,992 n/kg-s HM). [14] This suggests that the approaches for fuel fabrication that were proposed by the study should be investigated for the CORAIL-TRU multirecycling option. That study was supportive of the pyrochemical processing method because it is applicable in small facilities close to the reactor, as opposed to the aqueous processes that favor large facilities, requiring fuel shipment over long distances.

## 7.5 Helium Gas Production in Fuel Pin

The presence of Am-241 coupled with the relatively short half-life of Cm-242, leads to a buildup of helium gas during fuel pin irradiation. Fuels development experts have indicated that high concentrations of Am-241 in fabricated fuel pins are potentially problematic due to this source of helium production. Furthermore, during the core residence time of the assembly, there will be some helium production from the  $\alpha$ -decay of Cm-244 ( $T_{1/2}=18$  years) and Pu-238 ( $T_{1/2}=87.7$  years). These three nuclides have elevated values compared to the CORAIL-Pu case. An evaluation of the helium gas production rates in the CORAIL assembly has therefore been performed to assess the impact of these elevated levels. Results are summarized in Table 7.4, for the amounts of gaseous elements produced. More detailed analysis of the release rates out of the fuel is needed.

The amount of gaseous fission products is roughly constant for all cases because the average amount of fission energy generated by the assembly is the same. On the other hand, the amount of helium produced increases with the contents of the three nuclides. These results confirm that there is a direct relationship between higher actinide loading and helium production.

While the helium fraction of the total gas production is higher for the TRU recycling cases compared to the CORAIL-Pu cases, it is however relatively small ( $< 12\%$ ) in magnitude. It is only in the equilibrium recycle stage of the CORAIL-TRU option that it becomes about 20%. A solution would be required for containing any proportional increase in the release of the gases. The implications of such a design change on assembly handling and thermal-hydraulic performance should be analyzed. It is noted that PWR fuel pins release only a small fraction of their fission gas (about 1% or less). [17] This release level suggests that the slightly elevated level of gas production might not constitute additional problems. However, because of the preferential release of helium, this issue has to be additionally investigated.

## 7.6 Summary of Results

The results presented here show that the decay heat, gamma and neutron sources and doses at the separation and fabrication stages increase with recycling. This is attributed to the increase in the minor actinide content with recycling. The primary nuclide is Cm-244 which contributes to both the elevated decay heat and the significant increase in the neutron source. The build-up of Cf-252 with recycling adds to the neutron source level and actually becomes the leading contributor to it after the sixth recycle

stage. The major impact of the recycle of curium and higher actinides on the fuel cycle appears mainly at the fabrication stage, because of the elevated neutron source and dose.

If aqueous processing is utilized, the high decay heat loads must be taken into account when optimizing the process to keep the temperatures of the process stream from being too high. The protection of workers at the reprocessing, fabrication, and reactor plants is mostly complicated by the high neutron emission rates which accompany minor actinide recycling.

Significant research is needed to demonstrate the fuel manufacturing, burnup behavior, and fuels separation, issues which are made more difficult because of the high decay heat and neutron emissions.

Collectively, these results confirm that appropriate shielding and remote handling will be required for multirecycling of the TRU in an LWR fuel cycle. The neutron and gamma emission rates are however similar to or smaller than those that have been quoted for fast spectrum systems that would perform similar transmutation missions. [13,14] Therefore, *if remote handling of fuel is a cost that is acceptable* to the transmutation mission at least, *seven recycle* of the TRU appears possible. However, it should be noted that such a fuel cannot be supported by the current LWR technology. So additional work is required in this area to provide more definitive information on what can be done in current LWR fuel cycle. An option that has been proposed in this regard is to recycle the TRU three times in the LWR fuel cycle, and transfer the recovered TRU to a fast reactor system for ultimate consumption. In this approach, the fuel would be cooled for a long time prior to utilization in the fast reactor fuel cycle. This provides additional time for the deployment of the fast spectrum systems or advanced reactor systems.

Conceptually, developing infrastructure for a single recycle in the CORAIL assembly might not be beneficial (besides generating energy). This is because the concept is a delay line, and such a single recycle will result in a TRU vector that is more degraded than that in the legacy spent nuclear fuel, complicating subsequent transmutation. Additionally, the amount of material to be disposed after the single recycle, will be similar to that of the legacy waste.

**Table 7.1. Isotopic Composition for Primary Actinides at Charge Stage in TRU Multi-recycling.**

			UOX	MOX	CORAIL- Pu	CORAIL-TRU						
Cycle			-	-	7	2	3	4	5	6	7	Equilib.
U enrichment, %			4.0		4.57	4.85	4.98	5.02	5.03	5.04	5.04	5.12
TRU content, %				9.4	8.18	7.48	8.39	9.37	10.31	11.16	11.93	20.2
Fissile, %			4.0	63.63	46.84	49.37	45.23	42.72	41.20	40.13	39.31	32.83
U234			4.0	0.051	0.064							
U235				0.003	0.002							
U236				0.006	0.006							
U238			96.0									
Np237						1.76	3.00	3.17	3.13	3.04	2.93	1.82
Np239						0.00	0.00	0.00	0.00	0.00	0.00	0.00
Pu238				3.134	3.895	2.81	5.34	6.99	8.00	8.64	9.08	10.20
Pu239				56.346	36.056	38.40	34.17	32.27	31.05	30.16	29.47	24.40
Pu240				26.610	26.967	26.89	24.37	23.05	22.48	22.24	22.16	21.67
Pu241				7.283	10.784	10.80	9.84	9.14	8.72	8.44	8.24	7.13
Pu242				5.829	21.136	10.19	11.50	12.42	12.95	13.30	13.55	16.93
Am241				0.738	1.092	5.27	5.63	5.54	5.49	5.49	5.52	5.91
Am242m						0.02	0.03	0.04	0.04	0.04	0.05	0.07
Am243						2.41	3.28	3.58	3.72	3.81	3.86	4.59
Cm242						0.00	0.00	0.00	0.00	0.00	0.00	0.00
Cm243						0.01	0.03	0.03	0.03	0.03	0.03	0.03
Cm244						1.06	2.22	2.84	3.18	3.36	3.46	3.90
Cm245						0.14	0.43	0.63	0.76	0.85	0.90	1.14
Cm246						0.00	0.07	0.18	0.30	0.42	0.54	1.74
Cm247						0.00	0.00	0.01	0.02	0.02	0.03	0.15
Cm248						0.00	0.00	1.80E-03	5.40E-03	1.11E-02	1.88E-02	0.29
Bk249						0.00E+0	0.00E+0	0.00E+00	0.00E+00	0.00E+00	0.00E+00	3.00E-05
Cf249						0.00E+0	0.00E+0	6.00E-05	2.20E-04	5.00E-04	9.10E-04	1.91E-02
Cf250						0.00E+0	0.00E+0	1.00E-05	5.00E-05	1.00E-04	1.80E-04	3.48E-03
Cf251						0.00E+0	0.00E+0	1.00E-05	5.00E-05	1.20E-04	2.10E-04	4.95E-03
Cf252						0.00E+0	0.00E+0	0.00E+0	1.00E-05	2.00E-05	4.00E-05	9.40E-04
Cf253						0.00E+0	0.00E+0	0.00E+0	0.00E+0	0.00E+0	0.00E+0	0.00E+0
Es253						0.00E+0	0.00E+0	0.00E+0	0.00E+0	0.00E+0	0.00E+0	0.00E+0
Mass balance (kg per assembly )	Pu	Charge	0.0		13.8	11.7	12.9	14.2	15.5	16.6	17.7	28.7
		Discharge	6.0	N/A	14.6	13.4	14.7	15.9	17.1	18.1	19.1	29.3
		Net	6.0		0.8	1.7	1.7	1.7	1.6	1.5	1.4	0.6
	MA	Charge	0.0		0.2	1.4	2.2	2.7	3.1	3.4	3.7	7.0
		Discharge	0.5		1.4	1.8	2.3	2.7	3.1	3.4	3.7	6.7
		Net	0.5		1.2	0.4	0.2	0.1	0.0	0.0	-0.0	-0.3
	TRU	Charge	0.0		14.0	13.1	15.1	16.9	18.6	20.0	21.4	35.7
		Discharge	6.5		16.0	15.1	17.0	18.7	20.2	21.5	22.8	36.0
		Net	6.5		2.0	2.1	1.9	1.8	1.6	1.5	1.4	0.3

NOTE: For the MOX and CORAIL cases, the vectors are for the TRU content of MOX fuel pin; the MOX-pin heavy-metal mass additionally includes depleted uranium (80-90w/o).

**Table 7.2. Comparison of Key Radiation Parameters for CORAIL-TRU Assembly Recycle Stages.**

		UOX	MOX	CORAIL-Pu	CORAIL-TRU <sup>a)</sup>					
Cycle		-	-	7	2	3	4	5	6	7
Decay Heat (Watt)	Fabrication	0.01045	1983	2084	4337	9227	13540	16800	19240	21150
	Charge	0.01045	2054	684.5	1354	2888	4220	5226	5985	6580
	Discharge	2.06E+06	1.98E+06	2.09E+06	2.02E+06	2.01E+06	1.98E+06	1.98E+06	1.97E+06	1.96E+06
	After cooling	2515	5584	4138	5065	6468	7503	8295	8908	9502
Neutrons (n's/sec)	Fabrication	1.23E+04	9.48E+07	1.16E+08	9.68E+09	2.43E+10	4.39E+10	8.22E+10	1.52E+11	2.60E+11
	Charge	1.23E+04	9.69E+07	3.74E+07	2.94E+09	7.36E+09	1.26E+10	2.12E+10	3.58E+10	5.77E+10
	Discharge	1.23E+09	1.33E+10	8.61E+09	1.34E+10	2.71E+10	6.49E+10	1.43E+11	2.67E+11	4.51E+11
	After cooling	5.74E+08	6.47E+09	5.24E+09	7.97E+09	1.44E+10	2.69E+10	4.99E+10	8.53E+10	1.37E+11
Gamma (Watt)	Fabrication	0.00	0.56	0.60	3.45	5.58	7.26	8.44	9.42	10.29
	Charge	0.00	0.90	0.33	1.11	1.91	2.44	2.81	3.12	3.39
	Discharge	567015.04	522794.60	563810.66	549967.76	546426.92	539553.53	537839.18	534843.09	531462.47
	After cooling	1066.26	896.59	1035.50	1024.35	1021.37	1006.22	1000.93	995.13	989.32

a) The TRU vector used for estimating these parameters were obtained from TRANSEQM calculations (see Appendix K).

**NOTES:**

1. The fabrication stage data is for one metric ton of MOX pin. Charge data is based on one metric ton of heavy-metal (U and TRU) present in the charge assembly. Data for discharge and after cooling (five years) are based on the resulting mass after irradiation (~0.954 ton in all cases). The discharge and five year after discharge (after cooling) data include contributions from fission products.
2. Gamma source is expressed in equivalent MeV/s (or watts), as a means of providing indications of the relative unshielded gamma dose rates. This approach does not capture the overall gamma dose, however, particularly for shielded situations.

**Table 7.3. Comparison of Dose Rate of CORAIL MOX Fuel at Charge Stage (rem/hr).**

		CORAIL-Pu			CORAIL-TRU		
Cycle		1	7	Equilibrium <sup>a)</sup>	2	7 <sup>a)</sup>	Equilibrium <sup>a)</sup>
Surface of pellet	SF <sup>b)</sup>	0.006 ± 0.001	0.013 ± 0.001	0.014 ± 0.001	2.00 ± 0.001	38.00 ± 0.001	1039 ± 0.001
	( $\alpha$ ,n)	0.010 ± 0.001	0.018 ± 0.001	0.016 ± 0.001	0.038 ± 0.001	0.188 ± 0.001	0.34 ± 0.001
	Photon	3.004 ± 0.012	5.334 ± 0.019	4.893 ± 0.019	13.07 ± 0.016	57.06 ± 0.016	110.6 ± 0.015
	Total	3.02	5.36	4.92	15.11	95.25	1149.94
Surface of fuel cladding	SF <sup>b)</sup>	0.009 ± 0.003	0.018 ± 0.003	0.019 ± 0.003	2.73 ± 0.003	52.44 ± 0.003	1436 ± 0.003
	( $\alpha$ ,n)	0.013 ± 0.003	0.023 ± 0.003	0.021 ± 0.003	0.047 ± 0.003	0.24 ± 0.003	0.43 ± 0.003
	Photon	0.056 ± 0.139	0.101 ± 0.135	0.094 ± 0.134	1.68 ± 0.041	7.04 ± 0.041	18.46 ± 0.040
	Total	0.08	0.14	0.13	4.46	59.72	1454.89
One meter away from the surface of cladding	SF <sup>b)</sup>	2.7E-5 ± 0.002	5.6E-5 ± 0.003	6.1E-5 ± 0.003	0.01 ± 0.003	0.17 ± 0.003	4.61 ± 0.003
	( $\alpha$ ,n)	4.0E-5 ± 0.002	6.8E-5 ± 0.002	6.2E-5 ± 0.003	1.4E-4 ± 0.003	7.1E-4 ± 0.003	1.3E-3 ± 0.003
	Photon	1.7E-4 ± 0.101	3.1E-4 ± 0.001	3.0E-4 ± 0.099	0.01 ± 0.039	0.03 ± 0.040	0.06 ± 0.037
	Total	2.32E-04	4.31E-04	4.20E-04	0.02	0.20	4.67

a) Heavy metal isotope vector was calculated by the TRANSEQM code.

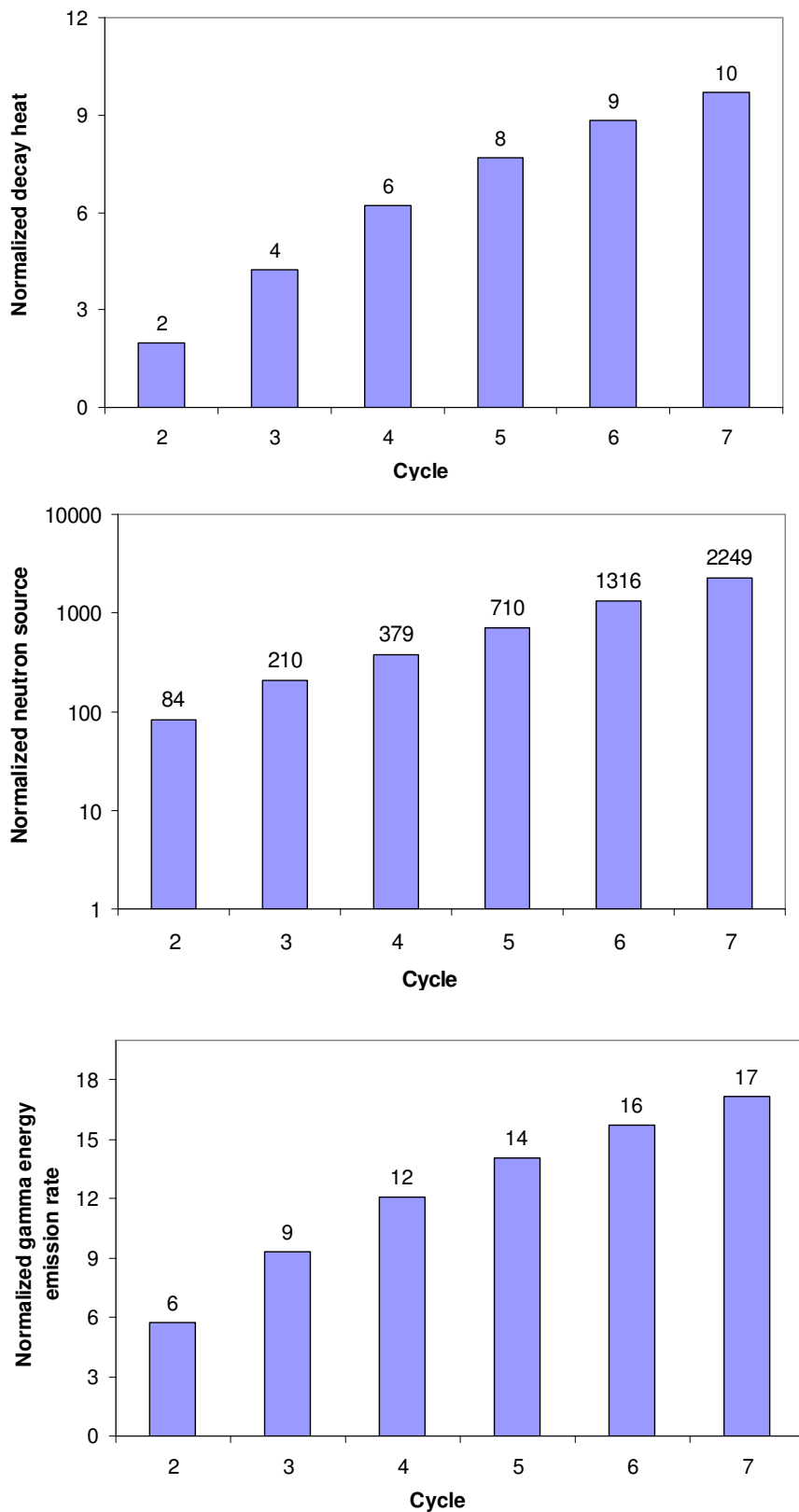
b) SF means spontaneous fission neutrons.

Note:

1. Flux-to-dose rate conversion factors for neutrons and photons are from ANSI/ANS-6.1.1-1977 (obtained from MCNP manual).
2. Spectrum for the spontaneous fission neutrons is from MCNP4 and that for photons comes from ORIGEN2 calculations.
3. Spectrum for ( $\alpha$ ,n) neutrons is based on generic data for MOX fuel (see Appendix E).
4. Number of neutron histories = 1.0E+06, photon = 5.0E+06

**Table 7.4. Comparison of Fission Gas Content of CORAIL Assemblies at Discharge (moles/assembly).**

	CORAIL-Pu			CORAIL-TRU		
	Cycle 1	Cycle 7	Equilibrium	Cycle 2	Cycle 7	Equilibrium
He	0.59	0.99	1.13	2.27	3.90	6.35
Kr	2.62	2.64	2.63	2.66	2.53	2.43
Xe	28.09	27.84	27.71	27.72	26.84	26.71
Total	31.29	31.47	31.46	32.65	33.27	35.49



**Fig. 7.1. Comparison of Fuel Handling Indices at Fabrication Stage of TRU Multirecycling (Compared to CORAIL-Pu)**

## 8.0 Indices for Proliferation Considerations

For proliferation considerations, the critical masses, heating rates and neutron sources consistent with the TRU vectors provided in Table 8.1, have been calculated and compared. These vectors are slightly different from those listed in Section 6.0 and 7.0. This is because the study discussed here were performed prior to the development of the TRANSEQM procedure and CORAIL-TRU vectors based on Method II of Ref. 3 were employed for the earlier study. Since the conclusions of this study are not expected to be different, these calculations have not been revised.

For the results presented here, it is assumed in that the plutonium or TRU and impurity uranium compositions exist in isolated forms during the separations campaign. This is an important consideration because if the plutonium or TRU could be co-processed with the depleted uranium required to make the MOX pin, the critical mass becomes quite large.

**Table 8.1. Isotopic Compositions for Primary Actinides in Various Fuel Forms.**

				TRU composition in MOX						
		UO <sub>2</sub>	W-Pu	MOX	CORAIL-Pu			CORAIL-TRU		
					Cycle 1	Cycle 2	Cycle 7	Cycle 1	Cycle 2	Cycle 7
U enrichment,%		4.00			4.15	4.40	4.57	4.50	4.62	4.86
Pu or TRU content in MOX pin, %			100.00	9.40	6.50	6.81	8.18	9.00	12.50	20.00
Charge TRU vector, %	Am241			0.738	0.700	1.196	1.092	4.654	5.685	6.476
	Am242m							0.019	0.035	0.081
	Am243							1.472	2.290	3.769
	Cm242							0.000	0.000	0.000
	Cm243							0.005	0.023	0.031
	Cm244							0.496	1.415	3.141
	Cm245							0.038	0.266	0.938
	Np237							6.641	4.405	2.006
	Pu238			3.134	2.700	3.060	3.895	2.749	6.549	11.003
	Pu239		93.4	56.346	56.000	42.838	36.056	48.652	36.768	27.264
	Pu240		6	26.610	25.900	30.029	26.967	22.980	24.842	23.666
	Pu241		0.6	7.283	7.400	11.807	10.784	6.926	9.669	7.668
	Pu242			5.829	7.300	11.012	21.136	5.033	7.791	13.699
	U234			0.051		0.050	0.064	0.000	0.107	0.180
	U235	4		0.003		0.002	0.002	0.002	0.003	0.002
	U236			0.006		0.006	0.006	0.002	0.006	0.005
	U238	96				0.000	0.000	0.325	0.146	0.072
Fissile		4.00	94.00	63.63	63.40	54.65	46.84	55.64	46.76	35.98

In addition to the critical masses for the  $\text{UO}_2$ , MOX, and CORAIL fuel, those for weapons-grade Pu and 20w/o enriched uranium fuel are also included for comparison. Uranium is classified as highly enriched when the U-235 (fissile) content is 20w/o or greater. The results in Table 8.2 show that the critical mass for weapons-grade Pu with a fissile content of 94 w/o is 10.6 kg. For the LWR spent nuclear fuel that has been recycled for six stages in the CORAIL-Pu and TRU assemblies, the fissile contents are 47w/o and 36w/o, respectively, and the critical masses are 17.9 and 17.5 kg. (The fuel vectors are different for these cases.) The critical mass of the 20w/o enriched uranium fuel is about 700 kg. It should be noted that the critical mass of the heavy-metal in a MOX pin (with 1-20w/o Pu or TRU and 80-99w/o depleted uranium) is greater than 3.5 tons. For some mixture of the MOX pin, a critical mass cannot be obtained in dry condition.

**Table 8.2. Critical Mass for Bare Sphere of Transuranics Only.**

Case	Fissile Content (%)	Critical Mass (kg)
Weapons-Grade Pu	94.0	10.6
Uranium (20w/o U-235)	20.0	701.4
CORAIL-Pu		
Stage 1	63.4	14.3
Stage 2	54.6	15.7
Stage 7	46.8	17.9
CORAIL-TRU		
Stage 1	55.6	15.4
Stage 2	46.8	16.2
Stage 7	36.0	17.5

Taken collectively, the results show that an isolated stream of plutonium and TRU fuel could be attractive to proliferators because of the relatively small amounts of material required to achieve a critical mass. The potential for using these materials for making nuclear bombs is however not fully reflected by the magnitude of the critical mass (though a primary indicator). The issues of decay heat and spontaneous neutron source also have to be considered. This is because they are important in estimating the potential yields of such weapons. The higher the decay heat and spontaneous neutron source, the more difficult it is to get the destructive performance from the nuclear device.

Estimates of the relative decay heat and neutron source from the CORAIL fuel to that of the weapons-grade Pu revealed that the decay heat of the fuel is a factor of 10-120 greater and the neutron source is a factor of 10-11,000 greater. The low end is for the cycle 1 fuel, while the high end is for the cycle 7 fuel. With these increased values, relative to weapons-grade Pu, it is clear that remote fabrication and assembly facilities and temperature-insensitive materials would be required to use the TRU or plutonium derived from the CORAIL concept for weapons purposes. Additionally, the separation hypothesis also has an impact on the attractiveness of the material. If the TRU or plutonium is not isolated, the critical mass becomes so large that attainment of the required amounts implies that a substantial investment has been made by the country producing the material. This should be readily detected by international organizations assigned to tracking the non-civil uses of nuclear materials.

## 9.0 Conclusions and Future Work

Preliminary evaluations of the indices for assessing fuel proliferation and handling concerns have been done for the CORAIL fuel assembly proposed for multirecycling of plutonium (Pu) or transuranics (TRU) in LWR fuel cycles. Different separation hypotheses were also investigated in order to assess various proliferation resistant fuel cycles for TRU multirecycling in PWRs. Both elemental and isotopic separation options were investigated. Data for different recycle stages and fuel cycle states of the CORAIL assemblies were intercompared to each other and to those for UO<sub>2</sub> and MOX fuel assemblies, and additionally to weapons-grade Pu fuel for proliferation considerations.

The recycling of neptunium in the fuel cycles provides incremental benefit to the proliferation resistance of the fuel cycle, because of the denaturing of the plutonium vectors (relative increase in Pu-238). However, the dose rates do not increase more than a factor of two over that of the Pu-only recycling case and both of these fuel forms are not self-protecting. The additional recycling of neptunium, while providing additional mass reduction, has minimal effect on the radiotoxicity, dose rate, and long-term heating rate, compared to the Pu-only case. So this option provides only marginal benefits.

Additionally recycling Am with Pu+Np provides significant benefits to the fuel repository, the fuel cycle, and the proliferation resistance of the fuel. Compared to fuel cycle results for the Pu-only case, the decay heat levels are increased a factor of 2 to 6, the neutron source/dose by a factor of 2 to 4, and the gamma dose by a factor of ~10 at the fabrication and charge states. The benefit provided by this approach is however limited if Cm-244 is buried in the repository, because of the decay of this nuclide to Pu-240, which dictates the intermediate term radiotoxicity and heat loads. The partial separation of plutonium and americium by isotopic separation, would help in this regard, as the amount of Cm-244 to be passed to the repository would be decreased. The fuel handling indices (radiation sources and decay heat) are however reduced, primarily because the absence of Pu-242 reduces the TRU content of the fabricated MOX pin. The separation of americium from curium however presents technological problems that have to be addressed. Additionally, isotopic separation on the scale that would support the transmutation mission could be quite expensive and make the partial separation option unattractive.

TRU recycling provides the most benefit to the repository, both in terms of the radiotoxicity and dose rate, and the heat load. While this option is also beneficial from

proliferation resistance viewpoint, its application would complicate fuel handling, and at the least make it most expensive, because of the shielding requirements. It would be useful to have a comparative analysis of this shielding cost to that of multirecycling of TRU in a fast reactor fuel cycle. It is anticipated that *remote fuel handling would be required in any event for TRU multirecycling in LWR systems.*

In regards to fuel handling issues, the preferred approach is to recycle plutonium, neptunium, and americium, and then store curium (taken as Cm-244 in this discussion) for a few of its half-lives. This provides both proliferation resistance and repository benefits. With this approach the radiotoxicity curve would be similar to that for the CORAIL-TRU case. If Cm-244 is stored for 360 years, which would be in the time scale of the period in which fission products dominate the decay curve, nearly all the Cm-244 would have decayed to Pu-240. Following this time, the Pu-240 could be burned as special targets in the nuclear fuel cycle. The modalities for Cm-244 storage outside the repository would have to be evaluated. However, because of its high specific neutron source value, such a storage facility will require sufficient shielding and result in added cost to the fuel cycle. Criticality and heat load concerns in the storage facility are additional issues that have to be addressed before this option becomes viable.

A cycle by cycle analysis of the CORAIL-TRU concept was conducted to investigate the practical limit to the number of recycle stage that could be tolerated before preventive measures makes the concept unworkable or expensive. It was observed that gamma and neutron sources and doses at the separation and fabrication stages increase with recycling. This is attributed to the increase in the minor actinide content with recycling. The primary nuclide is Cm-244, which contributes to both the elevated decay heat and the significant increase in the neutron source. The build-up of Cf-252 with recycling adds to the neutron source level and actually becomes the leading contributor to it after the sixth recycle stage. The major impact of the recycle of curium and higher actinides on the fuel cycle appears mainly at the fabrication stage, because of the elevated neutron source and dose.

If aqueous processing is utilized, the higher decay heat loads must be taken into account when optimizing the process to keep the temperatures of the process stream from being too high. The protection of workers at the reprocessing, fabrication, and reactor plants is mostly complicated by the high neutron emission rates which accompany minor actinide recycling. The results presented in this work indicate that the shielding requirement for the TRU multirecycling case is higher than that for the MOX-Pu or CORAIL-Pu options. If remote handling is a cost that is acceptable to the transmutation

mission, then at least seven recycles of the TRU can be performed in the CORAIL assembly. However, additional work is still required in this area to provide more definitive information on what can be done in current LWR fuel cycle.

The comparison of critical masses indicated that if the plutonium and TRU in the CORAIL MOX pins are isolated, the critical mass is less than 20 kg, which suggests that the fuel could be attractive to proliferants. The attractiveness level is however diminished by the relatively higher decay heat levels (10-120 times) and spontaneous neutron sources (10-11,000 times) compared to weapons-grade plutonium that has a fissile content of 94%. These indicators suggest complications in the handling of the material and the effectiveness of the nuclear device fashioned from the materials. However, the low critical mass might dictate that an advanced fuel separation concept be developed in which the plutonium and TRU are co-processed with the depleted uranium used in the fuel cycle. In this case, the critical mass would be quite large for the CORAIL fuel.

Some comparisons to a reference fast-reactor system might be necessary in the future for the usage, handling and radiotoxicity issues, particularly if it is logically assumed that the mission of TRU burning is better done in fast-spectrum systems. Preliminary evaluations [4] indicate that fast systems using fuel previously recycled in LWRs would also have to provide solutions to the fuel handling and usage issues, because of the relatively high minor actinide contents of the fuel. However, it is presumed that remote fuel handling and on-site processing will mitigate many of the safety implications associated with handling the transmuter fuel assemblies.

More detailed analyses of the dose rates and heating rates in the fuel cycle, and the required modifications to current technology, are planned in follow on studies. Indications of the shielding materials and dimensions required for worker protection and the temperatures corresponding to the heating rate will be determined. Cost analysis will also be performed to give indications of the expense associated with recycling TRU in LWRs.

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## APPENDIX A

### CORAIL Assembly Data

Parameter	Westinghouse 17x17
Number of rods (UO <sub>2</sub> /MOX)	180/84
Assembly pitch, cm	21.6112
Assembly gap, cm	0.1559
Fuel pitch, cm	1.26
Cladding material	Zr-4
Cladding outer radius, cm	0.474364
Cladding thickness, cm	0.0617
Cladding density, g/cm <sup>3</sup>	6.49012
Pellet radius, cm	0.41266
Pellet density g/cm <sup>3</sup>	10.02
Average coolant temperature, K	580
Coolant density, g/cm <sup>3</sup>	0.700594
Specific power density, W/g	36.055
Guide tube inner radius, cm	0.5715
Guide tube outer radius, cm	0.6120
Number of fuel assemblies	193
Power (thermal, electric), MW	3800/1300
System pressure, bar	155

## APPENDIX B

### Suggested Spent Fuel Heavy Metal Composition<sup>1</sup> for Commercial-Side ALWR: UO<sub>2</sub> Fuel, 4.2 w/o 235U, 50 GWd/t Burnup, 10 Years Cooling.

	<i>Weight Percent by Component</i>			
<b>Nuclide</b>	<b>Total HM</b>	<b>U</b>	<b>Pu</b>	<b>TRU</b>
U234	0.00007	0.020		
U235	0.00234	0.701		
U236	0.00194	0.585		
U238	0.32462	98.694		
Np237	6.64100			6.663
Pu238	2.74900		3.184	2.758
Pu239	48.65200		56.349	48.813
Pu240	22.98000		26.616	23.056
Pu241	6.92600		8.022	6.949
Pu242	5.03300		5.829	5.050
Am241	4.65400			4.669
Am242m	0.01900			0.019
Am243	1.47200			1.477
Cm242	0.00000			0.000
Cm243	0.00500			0.005
Cm244	0.49600			0.498
Cm245	0.03800			0.038
Cm246	0.00600			0.006

<sup>1</sup>99.995% U removed

## APPENDIX C

### One-Group Transmutation Code, TRANSEQM (T. K. Kim)

A one group transmutation code (called TRANSEQM) has been developed for searching for the equilibrium states of various CORAIL assembly concepts; TRANSEQM can also be used for discrete time calculations. The code solves the homogeneous, first-order ordinary differential equation,

$$\frac{d\bar{N}}{dt} = \mathbf{A}\bar{N}, \quad (1)$$

where  $\bar{N}$  is a  $M$ -dimensional vector of nuclide densities and  $\mathbf{A}$  is a  $(M \times M)$  transmutation matrix containing elements that are dependent on cross sections, decay constants and yield fractions. The transmutation chain that is considered by the code is displayed in Figure C.1. The solution of Eq. (1) is known for a given initial nuclide densities,  $\bar{N}(0)$ , and can be written as,

$$\bar{N}(t) = \left( \sum_{m=0}^{\infty} \frac{(\mathbf{A} \cdot \Delta t)^m}{m!} \right)^K \cdot \bar{N}(0), \quad (2)$$

where  $K$  denotes the total number of time intervals (for irradiation and/or decay), equivalent to the total time interval (i.e.,  $\Delta t = t/K$ ). In performing the summation indicated by Eq. (2), the norm of the transmutation matrix (i.e.,  $\|\mathbf{A}\| \cdot \Delta t$ ) is restricted to be less than  $\|\mathbf{A}\| \cdot \Delta t \leq 0.5$ , which limits the error to  $< 0.1\%$  if the first 4 to 5 terms of the summation indicated by Eq. (2) are employed. If  $\|\mathbf{A}\| \cdot \Delta t$  is greater than the limiting value, the specified norm is accomplished by repeatedly dividing the time by a factor of 2.

The TRANSEQM code searches the equilibrium state by utilizing two iteration steps. Figure C.2 shows the iteration scheme of the TRANSEQM code. TRU (or Pu) content and vector of the charge stage are determined with a given TRU content in the inner iteration and the uranium enrichment is determined in the outer iteration to maintain the desired fuel cycle length.

The accuracy of the TRANSEQM code was verified by comparing the results of the TRANSEQM and ORIGEN-RA codes. The results from this comparison are

summarized in Table C.1. The comparison is for fuel irradiated to a discharge burnup of 45,000 MWd/t and 5-year cooling after discharge. The two codes were supplied with the same fuel composition and one-group cross sections at the charged stage. Generally, the results of the TRANSEQM calculation agree well with those of the ORIGEN-RA calculation; the relative difference between the results of the one group transmutation solver and the ORIGEN-RA calculation are less than 0.1%. However, some isotopes (i.e., 0.26% of Pu-239, 0.93% of Cm-242, etc) are overestimated because the short lived isotopes are ignored in the transmutation chains of the one group transmutation equations.

**Table C.1. Comparison of Isotopic Mass (gram-atoms).**

	Charge	Discharge			After 5 year cooling after discharge		
		ORIGEN-RA	One group transmutation	Relative difference (%)	ORIGEN-RA	One group transmutation	Relative difference (%)
Th232	91.795	85.500	85.515	0.02	85.500	85.515	0.02
Pa233		0.186	0.186	0.00			
U233		3.500	3.500	0.00	3.685	3.686	0.03
U234		0.632	0.633	0.16	1.154	1.155	0.09
U235	164.892	70.480	70.482	0.00	70.490	70.489	0.00
U236		17.830	17.829	-0.01	17.840	17.842	0.01
U237		0.033	0.033	0.00			
U238	3788.833	3670.000	3670.657	0.02	3670.000	3670.657	0.02
Np237	3.657	3.652	3.647	-0.14	3.717	3.712	-0.13
Np239		0.279	0.279	0.00			
Pu238	13.105	13.100	13.116	0.12	13.220	13.238	0.14
Pu239	46.658	46.340	46.462	0.26	46.620	46.739	0.25
Pu240	27.166	24.080	24.068	-0.05	26.280	26.267	-0.05
Pu241	12.131	17.480	17.466	-0.08	13.790	13.779	-0.08
Pu242	20.721	20.770	20.735	-0.17	20.780	20.739	-0.20
Am241	7.436	2.186	2.185	-0.05	5.846	5.840	-0.10
Am242M	0.054	0.070	0.070	0.00	0.068	0.068	0.00
Am243	8.051	8.062	8.056	-0.07	8.060	8.052	-0.10
Cm242		0.639	0.645	0.93			
Cm243	0.042	0.049	0.049	0.00	0.044	0.044	0.00
Cm244	9.519	12.690	12.693	0.02	10.480	10.482	0.02
Cm245	2.306	2.322	2.322	0.00	2.321	2.321	0.00
Cm246	5.692	5.714	5.714	0.00	5.713	5.713	0.00
Cm247	0.453	0.454	0.454	0.00	0.454	0.454	0.00
Cm248	0.86	0.864	0.864	0.00	0.878	0.879	0.11
Bk249		0.025	0.025	0.00			
Cf249	0.042	0.018	0.018	0.00	0.042	0.042	0.00
Cf250	0.001	0.014	0.014	0.00	0.011	0.011	0.00
Cf251	0.001	0.010	0.010	0.00	0.010	0.010	0.00
Cf252	0.001	0.020	0.020	0.00	0.005	0.005	0.00

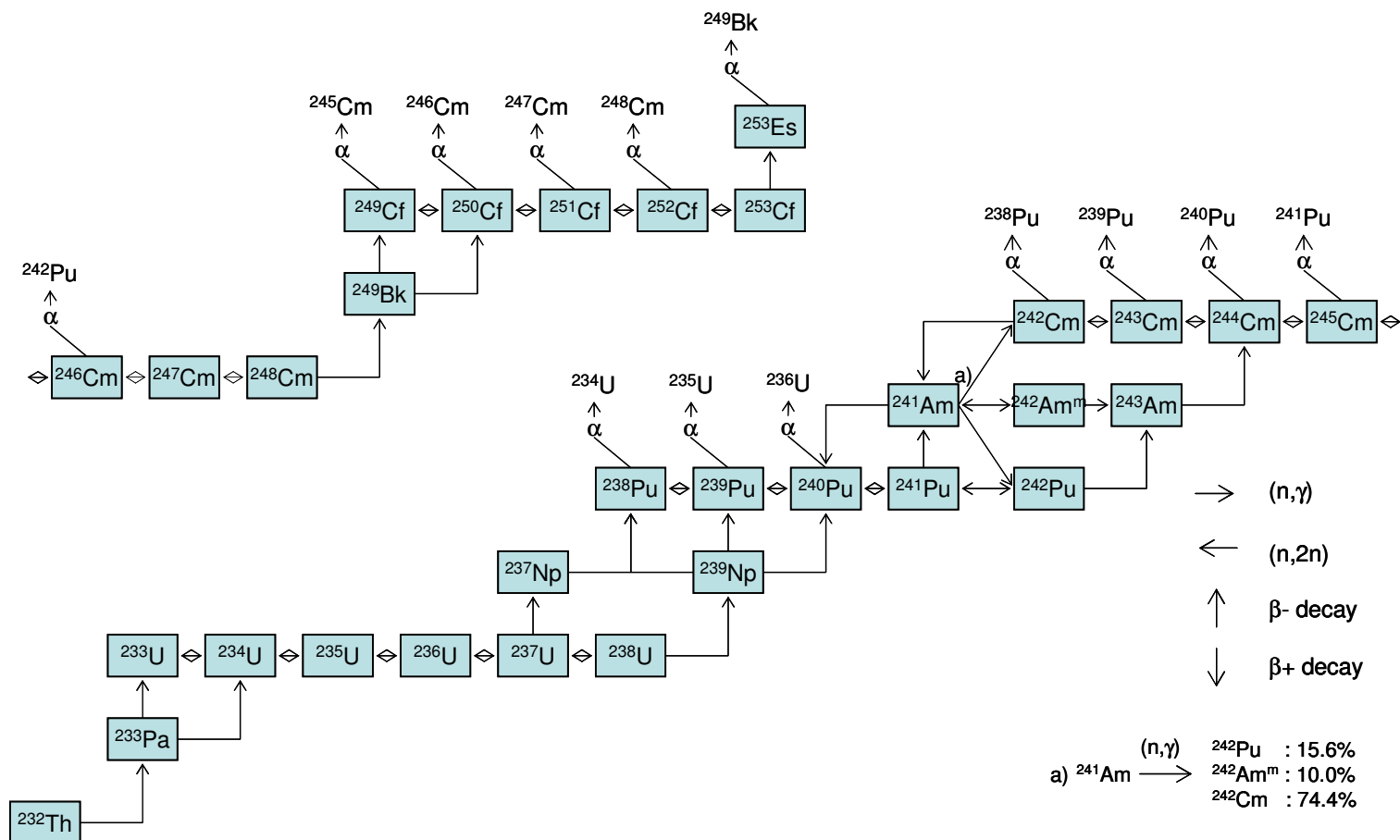
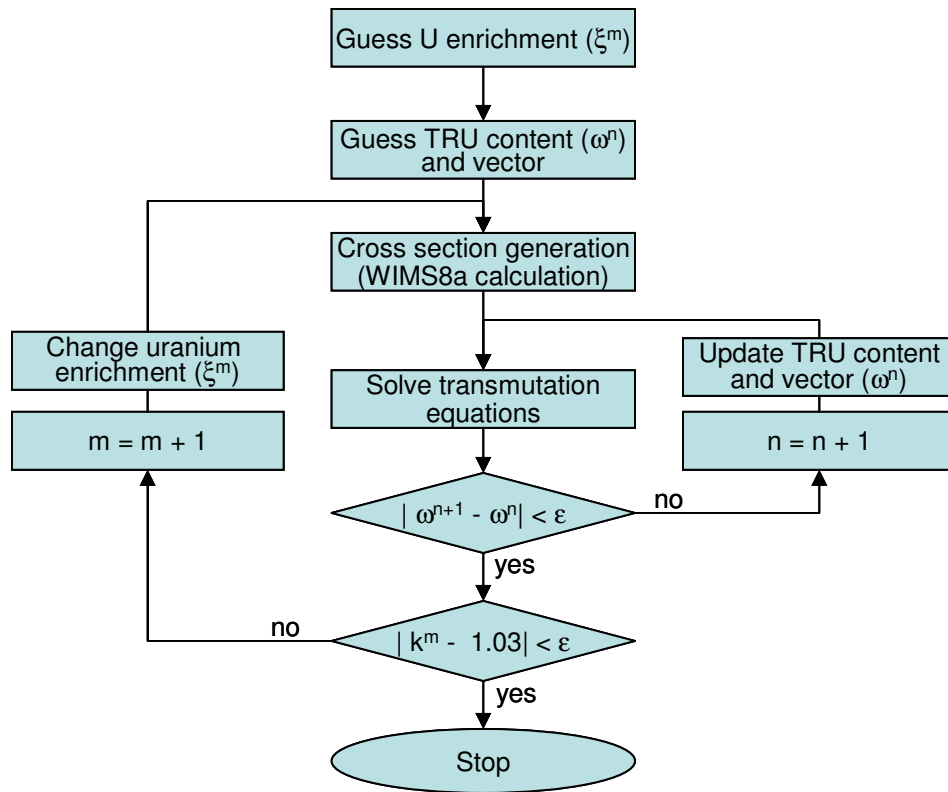


Fig. C.1. Transmutation Chain of Actinides in the TRANSEQM Code.



**Fig. C.2. TRANSEQM Equilibrium State Search Algorithm.**

## APPENDIX D

### Leading Contributions to Various Assessment Indices

**Table D.1. Leading contributors to the indices at fabrication stage of equilibrium states.**

	CORAIL-Pu		CORAIL-TRU		Pu-Np		Pu-Np-Am		No-Pu242		No Pu242 & Am243			
Cooling	5 year		5 year		20 year		5 year		5 year		5 year			
Decay heat (watt)	TOTAL	1960	TOTAL	38390	TOTAL	25520	TOTAL	3766	TOTAL	11600	TOTAL	8531	TOTAL	8138
	PU238	1731	CM244	24280	PU238	13020	PU238	3507	PU238	10210	PU238	7550	PU238	7218
	PU240	140	PU238	12000	CM244	9461	PU240	158	AM241	967	AM241	684	AM241	649
	PU239	55	AM241	1213	AM241	2338	PU239	63	PU240	236	PU240	174	PU240	165
	PU241	31	PU240	309	PU240	335	PU241	35	PU239	88	PU239	71	PU239	69
	PU242	3	CF252	124	CM243	128	PU242	3	AM243	52	PU241	38	PU241	37
			CM243	117	PU239	97			PU241	45	AM243	13		
			PU239	95	AM243	59			PU242	3				
			AM243	60	CM246	28								
			PU241	51	PU241	23								
			CM242	48	CF250	13								
	Neutrons (n's/sec)	TOTAL	1.20E+08	TOTAL	7.65E+12	TOTAL	2.45E+11	TOTAL	1.84E+08	TOTAL	4.60E+08	TOTAL	3.01E+08	TOTAL
PU238		5.78E+07	CF252	7.41E+12	CF252	1.22E+11	PU238	1.17E+08	PU238	3.41E+08	PU238	2.52E+08	PU238	2.41E+08
PU242		3.91E+07	CM244	9.62E+10	CM244	3.75E+10	PU242	4.10E+07	PU242	5.18E+07	PU240	2.66E+07	PU240	2.51E+07
PU240		2.14E+07	CF250	8.74E+10	CF250	3.63E+10	PU240	2.41E+07	PU240	3.60E+07	AM241	1.98E+07	AM241	1.87E+07
PU239		1.31E+06	CM246	3.16E+10	CM246	2.48E+10	PU239	1.50E+06	AM241	2.79E+07	PU239	1.69E+06	PU239	1.64E+06
U238		1.17E+04	CM248	2.61E+10	CM248	2.40E+10	U238	1.15E+04	PU239	2.07E+06	AM243	3.39E+05	U238	1.16E+04
U235		2.62E+00	PU238	4.01E+08	PU238	4.35E+08	NP237	1.40E+03	AM243	1.42E+06	U238	1.15E+04	AM242M	7.89E+03
			PU242	5.83E+07	AM241	6.75E+07	U235	2.57E+00	AM242M	1.54E+04	AM242M	8.61E+03	NP237	1.42E+03
			PU240	4.72E+07	PU242	5.64E+07			U238	1.07E+04	NP237	1.46E+03	U235	2.59E+00
			AM241	3.50E+07	PU240	5.11E+07			NP237	1.78E+03	U235	2.58E+00	PU242	1.98E-01
			CM242	1.03E+07	CM243	4.64E+06			U235	2.39E+00	PU242	1.99E-01	AM243	4.99E-03
Gamma (watt)	TOTAL	0.6	TOTAL	23.3	TOTAL	20.7	TOTAL	1.1	TOTAL	7.8	TOTAL	5.4	TOTAL	5.0
	PU238	0.5	CM244	6.3	AM241	10.2	PU238	1.0	AM241	4.2	AM241	3.0	AM241	2.8
			AM241	5.3	PU238	3.7			PU238	2.9	PU238	2.2	PU238	2.1
			CF252	4.1	CM243	2.7			AM243	0.5	AM243	0.1		
			PU238	3.4	CM244	2.4			PU240	0.1	PU240	0.1		
			CM243	2.5	AM243	0.6								
			AM243	0.6	CF249	0.5								
			CF249	0.6	CM245	0.1								
			CM245	0.2	PU240	0.1								
			PU240	0.1	CF252	0.1								

**Table D.2. Leading contributors to the indices at fabrication stage for TRU multi-recycling up to 7 cycle.**

Cycle	2		3		4		5		6		7			
Code	TRANSEQM		TRANSEQM		TRANSEQM.		TRANSEQM		TRANSEQM.		TRANSEQM		WIMS8	
Decay heat (watt)	TOTAL	4337	TOTAL	9227	TOTAL	13540	TOTAL	16800	TOTAL	19240	TOTAL	21150	TOTAL	21460
	CM244	2421	CM244	5894	CM244	8848	CM244	11050	CM244	12640	CM244	13830	CM244	13530
	PU238	1211	PU238	2548	PU238	3831	PU238	4822	PU238	5606	PU238	6260	PU238	6697
	AM241	451	AM241	481	AM241	525	AM241	563	AM241	605	AM241	650	AM241	809
	PU240	143	PU240	130	PU240	132	PU240	140	PU240	151	PU240	162	PU240	201
	PU239	55	PU239	55	PU239	58	PU239	62	PU239	65	PU239	68	CM243	74
	PU241	28	CM243	43	CM243	56	CM243	60	CM243	63	CM243	67	PU239	72
	CM243	15	PU241	32	PU241	32	PU241	33	PU241	34	PU241	35	PU241	37
	AM243	12	CM242	22	CM242	26	AM243	28	AM243	30	AM243	33	AM243	32
	PU242	1	AM243	19	AM243	24	CM242	27	CM242	28	CM242	30	CM245	7
CM245	1	CM245	2	CM245	4	CM245	5	CM245	6	CM246	7	PU242	2	
Neutrons (n's/sec)	TOTAL	9.67E+09	TOTAL	2.43E+10	TOTAL	4.39E+10	TOTAL	8.22E+10	TOTAL	1.52E+11	TOTAL	2.60E+11	TOTAL	5.39E+10
	CM244	9.58E+09	CM244	2.33E+10	CM244	3.50E+10	CM244	4.37E+10	CF252	9.55E+10	CF252	1.96E+11	CM244	5.36E+10
	PU238	4.04E+07	CM246	5.57E+08	CF252	6.84E+09	CF252	3.44E+10	CM244	5.00E+10	CM244	5.48E+10	PU238	2.24E+08
	PU240	2.17E+07	CF252	2.70E+08	CM246	1.60E+09	CM246	2.97E+09	CM246	4.48E+09	CM246	6.05E+09	PU240	3.07E+07
	AM241	1.30E+07	PU238	8.50E+07	CF250	1.51E+08	CF250	5.86E+08	CF250	1.43E+09	CF250	2.74E+09	PU242	2.93E+07
	PU242	1.28E+07	PU240	1.97E+07	PU238	1.27E+08	CM248	2.46E+08	CM248	5.54E+08	CM248	1.00E+09	AM241	2.34E+07
	PU239	1.30E+06	PU242	1.78E+07	CM248	7.43E+07	PU238	1.61E+08	PU238	1.87E+08	PU238	2.09E+08	CM243	2.67E+06
	CM243	5.36E+05	AM241	1.38E+07	PU242	2.17E+07	PU242	2.48E+07	PU242	2.74E+07	PU242	2.98E+07	PU239	1.71E+06
	AM243	3.12E+05	CF250	1.19E+07	PU240	2.01E+07	PU240	2.14E+07	PU240	2.30E+07	PU240	2.47E+07	AM243	8.57E+05
	CM245	1.71E+04	CM248	8.79E+06	AM241	1.51E+07	AM241	1.62E+07	AM241	1.74E+07	AM241	1.88E+07	CM245	1.90E+05
U238	1.18E+04	CM242	4.68E+06	CM242	5.45E+06	CM242	5.71E+06	CM242	5.98E+06	CM242	6.30E+06	U238	1.11E+04	
Gamma (watt)	TOTAL	3.4	TOTAL	5.6	TOTAL	7.3	TOTAL	8.4	TOTAL	9.4	TOTAL	10.3	TOTAL	11.0
	AM241	2.0	AM241	2.1	AM241	2.3	CM244	2.9	CM244	3.3	CM244	3.6	AM241	3.5
	CM244	0.6	CM244	1.5	CM244	2.3	AM241	2.5	AM241	2.7	AM241	2.8	CM244	3.5
	PU238	0.3	CM243	0.9	CM243	1.2	PU238	1.4	PU238	1.6	PU238	1.8	PU238	1.9
	CM243	0.3	PU238	0.7	PU238	1.1	CM243	1.3	CM243	1.4	CM243	1.4	CM243	1.6
	AM243	0.1	AM243	0.2	AM243	0.2	AM243	0.3	AM243	0.3	AM243	0.3	AM243	0.3
					CM245	0.1	CM245	0.1	CM245	0.1	CF252	0.1	CM245	0.1
										CM245	0.1	PU240	0.1	

**Table D.3. Leading contributors to the indices at charge stage for TRU multi-recycling up to 7 cycle.**

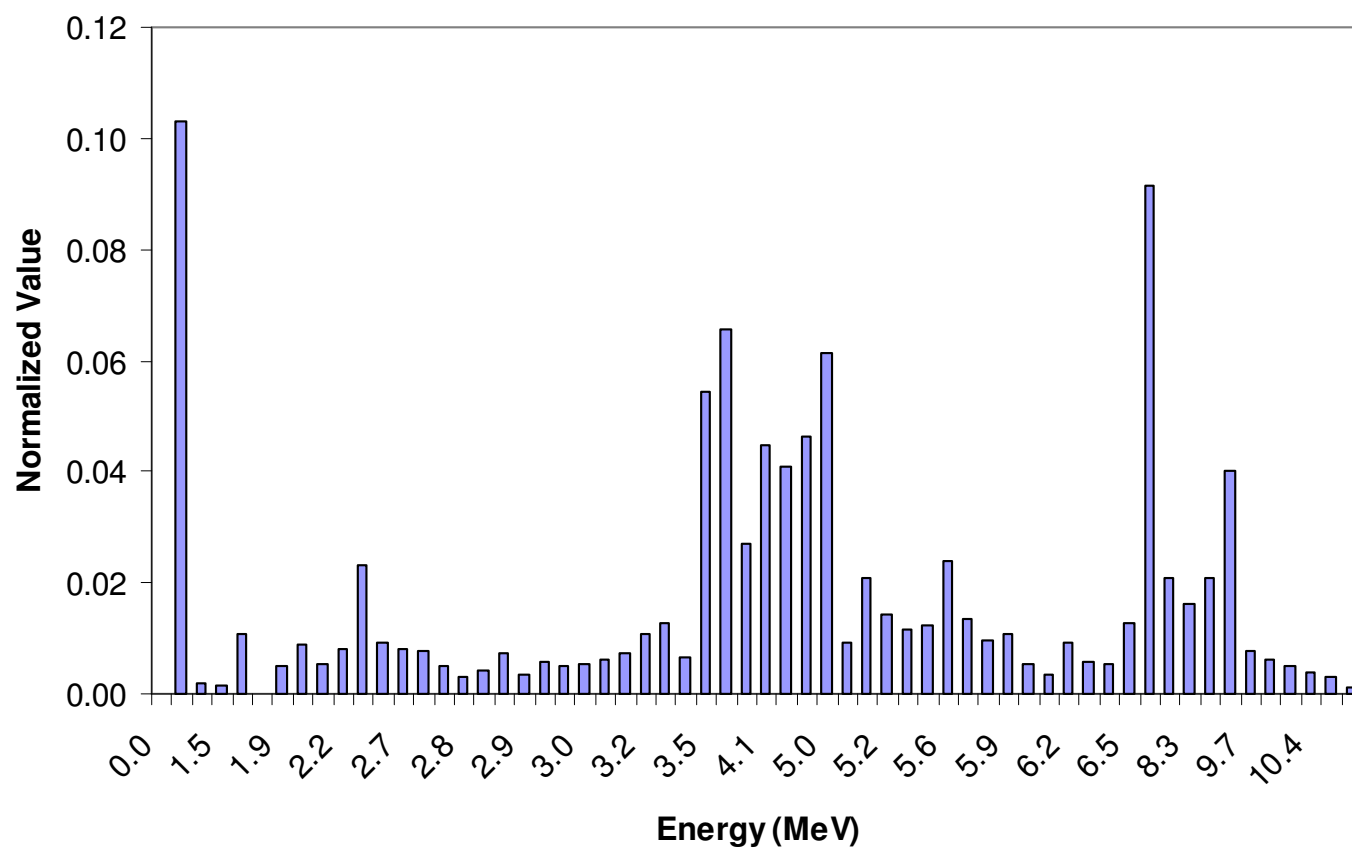
Cycle	2		3		4		5		6		7			
Code	TRANSEQM		TRANSEQM		TRANSEQM.		TRANSEQM		TRANSEQM.		TRANSEQM		WIMS	
Decay heat (watt)	TOTAL	1354	TOTAL	2888	TOTAL	4220	TOTAL	5226	TOTAL	5985	TOTAL	6580	TOTAL	6498
	CM244	734	CM244	1786	CM244	2681	CM244	3347	CM244	3830	CM244	4190	CM244	4002
	PU238	390	PU238	821	PU238	1234	PU238	1553	PU238	1805	PU238	2016	PU238	2105
	AM241	147	AM241	190	AM241	205	AM241	218	AM241	233	AM241	249	AM241	257
	PU240	47	PU240	43	PU240	44	PU240	47	PU240	50	PU240	54	PU240	64
	PU239	18	PU239	18	PU239	19	PU239	20	PU239	21	PU239	22	PU239	23
	PU241	8	CM243	14	CM243	18	CM243	19	CM243	20	CM243	21	CM243	22
	AM243	5	PU241	9	PU241	9	PU241	10	PU241	10	AM243	11	PU241	11
	CM243	4	AM243	6	AM243	8	AM243	9	AM243	10	PU241	10	AM243	10
	PU242	0	CM245	1	CM245	1	CM245	2	CM245	2	CM246	2	CM245	2
CM245	0	PU242	0	CM246	1	CM246	1	CM246	2	CM245	2	PU242	1	
Neutrons (n/sec)	TOTAL	2.94E+09	TOTAL	7.36E+09	TOTAL	1.26E+10	TOTAL	2.12E+10	TOTAL	3.58E+10	TOTAL	5.77E+10	TOTAL	1.60E+10
	CM244	2.91E+09	CM244	7.07E+09	CM244	1.06E+10	CM244	1.33E+10	CF252	1.85E+10	CF252	3.79E+10	CM244	1.59E+10
	PU238	1.30E+07	CM246	1.82E+08	PU238	1.33E+09	CF252	6.67E+09	CM244	1.52E+10	CM244	1.66E+10	PU238	7.03E+07
	PU240	7.13E+06	CF252	5.24E+07	AM241	5.26E+08	CM246	9.72E+08	CM246	1.47E+09	CM246	1.98E+09	PU240	9.81E+06
	AM241	4.25E+06	PU238	2.74E+07	PU240	4.44E+07	CF250	1.73E+08	CF250	4.22E+08	CF250	8.05E+08	PU242	9.35E+06
	PU242	4.21E+06	PU240	6.52E+06	PU239	4.12E+07	CM248	8.09E+07	CM248	1.82E+08	CM248	3.28E+08	AM241	7.43E+06
	PU239	4.26E+05	PU242	5.85E+06	CM243	2.43E+07	PU238	5.19E+07	PU238	6.03E+07	PU238	6.73E+07	CM243	8.11E+05
	CM243	1.68E+05	AM241	5.49E+06	AM243	7.12E+06	PU242	8.12E+06	PU242	8.98E+06	PU242	9.74E+06	PU239	5.46E+05
	AM243	1.02E+05	CF250	3.52E+06	PU242	6.66E+06	PU240	7.11E+06	PU240	7.65E+06	PU240	8.19E+06	AM243	2.74E+05
	U238	1.21E+04	CM248	2.88E+06	CM245	5.91E+06	AM241	6.30E+06	AM241	6.74E+06	AM241	7.20E+06	CM245	6.05E+04
CM245	5.62E+03	CM243	4.94E+05	NP237	6.35E+05	CM243	6.85E+05	CM243	7.21E+05	CM243	7.59E+05	U238	1.18E+04	
Gamma (watt)	TOTAL	1.1	TOTAL	1.9	TOTAL	2.4	TOTAL	2.8	TOTAL	3.1	TOTAL	3.4	TOTAL	3.4
	AM241	0.6	AM241	0.8	AM241	0.9	AM241	1.0	AM241	1.0	AM241	1.1	AM241	1.1
	CM244	0.2	CM244	0.5	CM244	0.7	CM244	0.9	CM244	1.0	CM244	1.1	CM244	1.0
	PU238	0.1	CM243	0.3	CM243	0.4	PU238	0.4	PU238	0.5	PU238	0.6	PU238	0.6
	CM243	0.1	PU238	0.2	PU238	0.4	CM243	0.4	CM243	0.4	CM243	0.4	CM243	0.5
	AM243	0.0	AM243	0.1	AM243	0.1	AM243	0.1	AM243	0.1	AM243	0.1	AM243	0.1
	PU240	0.0	PU240	0.0	CM245	0.0	CM245	0.0	CM245	0.0	CM245	0.0	CM245	0.0
	PU241	0.0	CM245	0.0	PU240	0.0	PU240	0.0	PU240	0.0	CF252	0.0	PU240	0.0
	CM245	0.0	PU239	0.0	PU239	0.0	CF252	0.0	CF252	0.0	PU240	0.0	PU239	0.0
	PU239	0.0	NP237	0.0	CF252	0.0	PU239	0.0	PU239	0.0	CF249	0.0	NP237	0.0

**Table D.4. Leading contributors to the indices at charge stage of equilibrium states.**

	CORAIL-Pu		CORAIL-TRU			Pu-Np		Pu-Np-Am		No-Pu242		No Pu242 & Am243		
Cooling	5 year		5 year		20 year		5 year		5 year		5 year		5 year	
Decay heat (watt)	TOTAL	655	TOTAL	11930	TOTAL	8278	TOTAL	1249	TOTAL	3788	TOTAL	2791	TOTAL	2662
	PU238	549	CM244	7356	PU238	4193	PU238	1129	PU238	3288	PU238	2432	PU238	2324
	PU240	46	PU238	3863	CM244	2865	PU240	52	AM241	362	AM241	263	AM241	251
	AM241	33	AM241	449	AM241	786	AM241	37	PU240	77	PU240	57	PU240	54
	PU239	18	PU240	103	CF253	191	PU239	21	PU239	29	PU239	23	PU239	23
	PU241	9	CM243	37	PU240	110	PU241	10	AM243	17	PU241	11	PU241	11
	PU242	1	PU239	31	CM243	40	PU242	1	PU241	13	AM243	4	AM242M	0
			AM243	24	PU239	32	NP237	0	PU242	1	AM242M	0	NP237	0
			PU241	20	AM243	19	U238	0	AM242M	0	NP237	0	U238	0
			CM246	15	ES253	14	U235	0	NP237	0	U238	0	U235	0
		CM245	12	CM246	9	PB210	0	U238	0	U235	0	AM243	0	
Neutrons (n's/sec)	TOTAL	3.96E+07	TOTAL	1.51E+12	TOTAL	6.22E+10	TOTAL	6.05E+07	TOTAL	1.50E+08	TOTAL	9.82E+07	TOTAL	9.36E+07
	PU238	1.83E+07	CF252	1.44E+12	CF252	2.37E+10	PU238	3.77E+07	PU238	1.10E+08	PU238	8.12E+07	PU238	7.76E+07
	PU242	1.29E+07	CM244	2.91E+10	CM244	1.14E+10	PU242	1.34E+07	PU242	1.70E+07	PU240	8.70E+06	PU240	8.21E+06
	PU240	6.98E+06	CF250	2.57E+10	CF250	1.07E+10	PU240	7.88E+06	PU240	1.18E+07	AM241	7.60E+06	AM241	7.24E+06
	AM241	9.46E+05	CM246	1.03E+10	CM246	8.12E+09	AM241	1.06E+06	AM241	1.05E+07	PU239	5.52E+05	PU239	5.37E+05
	PU239	4.28E+05	CM248	8.56E+09	CM248	7.84E+09	PU239	4.89E+05	PU239	6.78E+05	AM243	1.11E+05	U238	1.20E+04
	U238	1.20E+04	PU238	1.29E+08	ES253	3.07E+08	U238	1.20E+04	AM243	4.64E+05	U238	1.20E+04	AM242M	2.56E+03
	U235	3.64E+01	PU242	1.91E+07	PU238	1.40E+08	NP237	4.58E+02	U238	1.16E+04	AM242M	2.79E+03	NP237	4.68E+02
	NP237	2.28E-01	PU240	1.57E+07	AM241	2.27E+07	U235	3.85E+01	AM242M	4.99E+03	NP237	4.81E+02	U235	3.70E+01
	PB209	0.00E+00	AM241	1.30E+07	PU242	1.84E+07	PB209	0.00E+00	NP237	5.87E+02	U235	3.74E+01	PU242	6.49E-02
PB210	0.00E+00	CM243	1.33E+06	PU240	1.68E+07	PB210	0.00E+00	U235	4.31E+01	PU242	6.50E-02	AM243	1.63E-03	
Gamma (watt)	TOTAL	0.3	TOTAL	7.1	TOTAL	9.1	TOTAL	0.5	TOTAL	2.7	TOTAL	1.9	TOTAL	1.8
	PU238	0.2	AM241	2.0	AM241	3.4	PU238	0.3	AM241	1.6	AM241	1.2	AM241	1.1
	AM241	0.1	CM244	1.9	CF253	2.4	AM241	0.2	PU238	0.9	PU238	0.7	PU238	0.7
	PU240	0.0	PU238	1.1	PU238	1.2	PU240	0.0	AM243	0.2	AM243	0.0	PU240	0.0
	PU241	0.0	CF252	0.8	CM243	0.9	PU241	0.0	PU240	0.0	PU240	0.0	PU241	0.0
	PU239	0.0	CM243	0.8	CM244	0.7	PU239	0.0	PU239	0.0	PU241	0.0	PU239	0.0
	PU242	0.0	AM243	0.2	AM243	0.2	PU242	0.0	PU242	0.0	PU239	0.0	NP237	0.0
	U235	0.0	CF249	0.2	CF249	0.2	NP237	0.0	AM242M	0.0	NP237	0.0	AM242M	0.0
	U238	0.0	CM245	0.1	CM245	0.0	U235	0.0	NP237	0.0	AM242M	0.0	U235	0.0
	NP237	0.0	PU240	0.0	PU240	0.0	U238	0.0	U235	0.0	U235	0.0	U238	0.0
PB210	0.0	CF250	0.0	CF252	0.0	PB210	0.0	U238	0.0	U238	0.0	PU242	0.0	

## APPENDIX E

( $\alpha$ ,n) Spectrum for Dose Calculations



## APPENDIX F

### ASSESSMENT OF DOSE RATES FOR PU+NP FUEL – Impact on Proliferation Resistance of LWR Fuel (Hill, Taiwo, Salvatores)

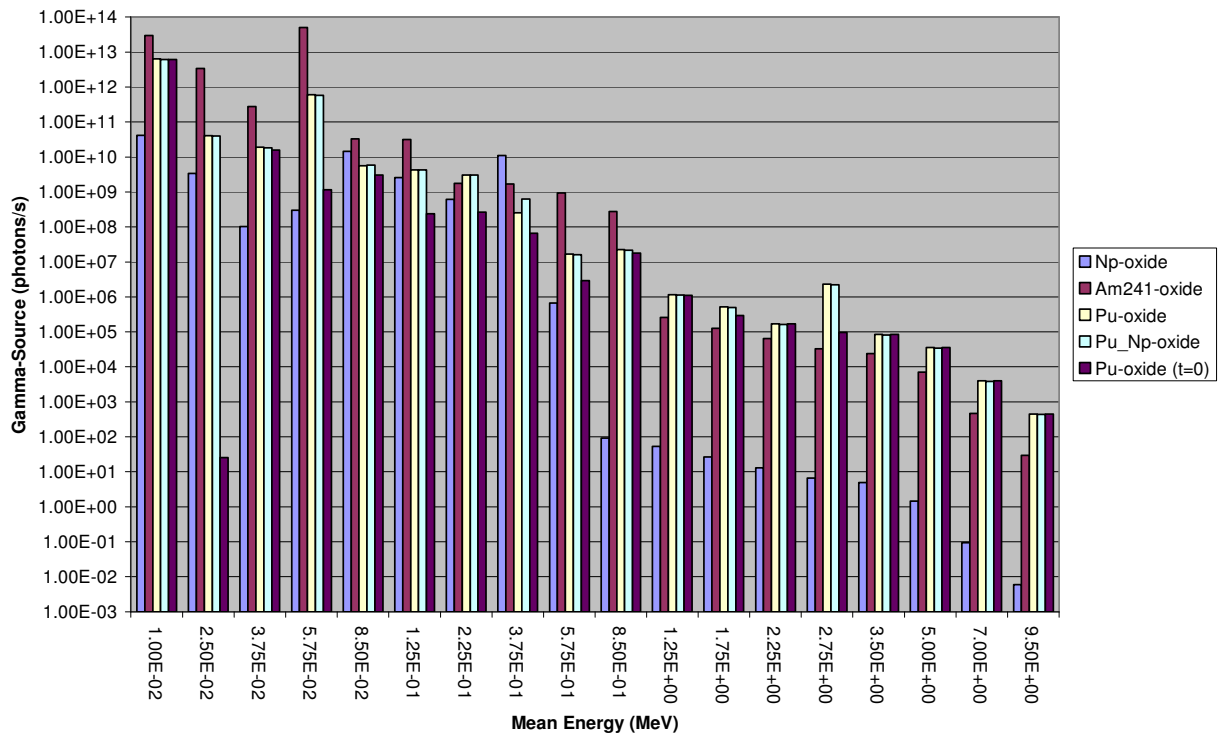
It has been suggested that the presence of neptunium-237 (Np-237) in the MOX (plutonium) fuel that is proposed for use in LWR-based transmutation systems would provide gamma sources that could enhance the proliferation resistance of the fuel. This idea came from the observation that adequate shielding is required for Np-237 powder that is being handled in some of the U.S. nuclear laboratories. The Np-237 sample decays via  $\alpha$ -emission to give protactinium-233 (Pa-233). The  $\beta$ -decay of the short-lived Pa-233 (half-life of 27 days) results in the emission of gammas (0.3 MeV), which are more energetic than those from reactor-grade plutonium decay chains. Therefore, the impact of the Pa-233 gammas depends highly on the level of shielding applied to the fuel. In order to assess this impact on proliferation resistance of recycled LWR fuel, calculations have been performed using the ORIGEN2 and MCNP codes.

The impact of gammas only has been evaluated in the current study. This is because in previous "proliferation resistance" evaluations of fuel cycle technologies, it was observed that the inclusion of minor actinides with the plutonium leads to an increase in the heat load, neutron source, and gamma source making the material less attractive for weapons utilization. The main contributors to heat load are Pu-238, Cm-244, and Am-241; whereas, the neutron source comes mainly from the curium and californium isotopes. Therefore, the only possible impact of Np would be for the gamma source. A more comprehensive evaluation of dose would however include that from neutrons.

For various heavy-metal oxides (Np-oxide, Pu-only-oxide, Pu+Np-oxide Am241-oxide), ORIGEN2 calculations were performed for a 1.056 kg quantity of heavy-metal, to determine the gamma source distribution. (This mass was selected to be consistent with that contained in a transmittal to ANL from ORNL on Np handling and was retained as it provides a basis for consistent comparison and normalization of data.) For the Np-oxide, Pu-only-oxide, Pu+Np-oxide cases, time-dependent ORIGEN2 decay calculations were performed. This is particularly important for the Np-237 case, as time is required for Pa-233 to reach its maximum activity state (after about 20 weeks); at that time, the activity from Np-237 and Pa-233 are about the same. It is noted that the ORIGEN2 code and hand-calculations (separately) yield the same activity (0.7 Curies/Kg) as contained in the

ORNL transmittal. For the Am-241 case, the gamma source distribution for the initial state only was determined. The ORIGEN2-calculated gamma source distributions for the cases are displayed in Fig. F.1.

The total gamma sources for the Am-241 and Pu-only cases were found to be respectively 1000 and 100 times higher than that for the Np-237 case. The Np-237 case has its highest gamma sources in the energy ranges with mean values of 0.01, 0.085 and 0.375 MeV, which correspond to the predominant gamma energies from Np-237 (0.014 and 0.086 MeV) and Pa-233 (0.098 and 312 MeV). For the Am-241 case, the predominant energies are 0.014 MeV and 0.06 MeV. It is important to note however that even for this case, the magnitude of the gammas in the group with a mean energy of 0.375 MeV is 15% of that for the Np-237 case. Additionally, at higher energies (importantly at 0.575 MeV), the gamma population for the Am-241 is much higher than that for the Np-237 case.



**Fig. F.1. Gamma Source Distribution (1.056 Kg of Material).**

For a given nuclear material type, the gamma source distribution generated by ORIGEN2 is used as the input source in an MCNP calculation. This calculation is done for 1.056 kg quantity of heavy-metal, with or without steel (Cr16Ni11Mo), which represents a container for the material. A simplified spherical geometry is assumed in the

calculation. The density of metal oxide is assumed to be 10 g/cc and the heavy-metal mass fraction in the oxide is estimated to be 88.1%. This gives a radius of 3.06 cm. Typically, 5 million starting particles were used in the MCNP calculations, but the low dose cases (<1 rem/hr) required 20 million particles to give decent statistics. The ANSI/ANS-6.1.1-1977 photon flux-to-dose rate conversion factors were employed for converting the surface photon flux to contact dose rate. The MCNP results are summarized in Table F.1. According to the transmittal from ORNL, for Np-237-oxide (1.056 kg Np-237) in a cylindrical container, the surface dose is ~ 5 to 6 rem/hr. The Np-237 oxide data on Table F.1 indicate that a steel thickness of about 5 cm is required to obtain a similar dose for the spherical configuration.

**Table F.1. Comparison of Gamma Dose Rates for Different Oxides with 1.056 kg Heavy-Metal.**

Case	Time (yr)	Steel thickness (cm)	Gamma dose (rem/hr)
<i>Np-237 oxide</i>	2	0.0	<b>17.2 (±0.5%)</b>
	2	0.1	12.0 (±0.4%)
	2	0.2	9.8 (±0.4%)
	2	0.3	8.3 (±0.4%)
	2	0.4	7.2 (±0.4%)
	2	0.5	6.3 (±0.4%)
	2	1.0	3.3 (±0.5%)
<i>Am-241 oxide</i>	0	0.0	<b>1424 (±2.0%)</b>
	0	0.5	4.9 (±8.5%)
	0	1.0	2.4 (±12%)
<i>Reactor Grade (RG) Pu oxide</i>	0	0.5	0.2 (±14%)
	2	0.0	<b>78.3 (±4.9%)</b>
	2	0.5	0.7 (±6.5%)
	2	1.0	0.3 (±8%)
<i>RGPu+Np oxide (~3.5%Np)</i>	2	0.5	0.9 (±6%)

The surface (contact) doses for spheres of Np-oxide, reactor-grade Pu oxide, and Am-241 oxide are 17 rem/hr, 78 rem/hr, and 1424 rem/hr, respectively. These results roughly reflect the trend in gamma energy source between the materials, with the Am being much hotter, then Pu, and then Np. The trend is also confirmed by the French

SUPERFACT experiment where pellet surface and fuel pin (very thin wall) dose rates were measured for standard MOX fuel pins (24% Pu enrichment) with 2% Np and 2% Am added (see Table F.2).

**Table F.2. Gamma Dose Rates for Two MOX Fuel Types Evaluated in the French SUPERFACT Experiment. (Ref. 18)**

Pin type	Pellet	Pin
2% Np	11.1 mGy/s-kg	6 mGy/s-kg
2% Am	44.4 mGy/s-kg	25 mGy/s-kg

These results confirm that the contact dose rate is a factor of four higher with the Am-241 added to the fuel, relative to a base case with Np-237. Therefore, the inclusion of Np-237 will not effectively increase the surface (contact) dose rate, especially as compared to Am-241.

The impact of steel (thin wall) thickness ranging from 0 to 1 cm was also evaluated for the same three materials discussed above (see Table F.1). The thin wall is particularly effective in shielding the reactor grade plutonium gammas, which are nearly all low energy. Whereas, the Pa-233 gamma (312 KeV) is harder to shield. If one mixed the Pu and Np at their ratios in 10 year cooled fuel (93%/7%), the contact dose would slightly decrease (74 rem/hr) as compared to separated plutonium, but with a 1 cm steel wall, the dose would increase from 0.3 rem/hr to 0.5 rem/hr.

In conclusion, mixing of the neptunium with the plutonium will not increase the contact gamma dose of the transuranic separation product, but will make the gamma dose harder to shield.

Regarding the gamma source, it is important to note that the fission products provide the "self-protection" of LWR spent fuel. For example, 1 MT of LWR spent fuel contains 100,000 Ci of Cs-137, but only 0.37 Ci of Np-237 (and Pa-233); and the Cs-137 has a higher energy gamma also. The Cs produces the 1,000 rem/hr at 1 meter at 30 years cooling. Thus, the gamma dose rates of the transuranic mixture will not approach the spent fuel standard, but should be viewed as a much smaller, but intrinsic barrier of the separated product.

## APPENDIX G

### GAMMA DOSE RATES FROM HEAVY METAL NUCLIDES WITH SIGNIFICANT CONTRIBUTIONS TO THE GAMMA SOURCE IN THE CORAIL FUEL CYCLE PU+NP FUEL

Using the calculation approach discussed in Appendix F, the gamma dose rates have been evaluated for the various nuclides with the highest contributions to the gamma source. The calculations were done for contact gamma dose on the surface of a material oxide containing 1.056 kg of a given transuranics or on the surface of an encasing 0.5 cm steel “shield” surrounding the material. Results from these cases are summarized in Table G.1.

**Table G.1. Comparison of Gamma Dose Rates for Different Oxides with 1.056 kg Heavy-Metal. --- Leading Actinide Contributors**

Case	Gamma dose (rem/hr)	
	Bare	Steel thickness of 0.5 cm
<i>Np-237 (t=2yrs)</i>	17.2 ( $\pm 0.5\%$ )	6.3 ( $\pm 0.4\%$ )
<i>Pu-238</i>	1036 ( $\pm 5.5\%$ )	2.0 ( $\pm 17\%$ )
<i>Pu-239</i>	1.6 ( $\pm 4.7\%$ )	0.1 ( $\pm 2.5\%$ )
<i>Pu-240</i>	13.2 ( $\pm 5.5\%$ )	0.04 ( $\pm 16\%$ )
<i>Pu-241 (t=0yrs)</i>	15.0 ( $\pm 5.6\%$ )	0.0 ( $\pm 0.0\%$ )
<i>Pu-241 (t=2yrs)</i>	150.9 ( $\pm 5.6\%$ )	3.6 ( $\pm 5.4\%$ )
<i>Am-241</i>	1424 ( $\pm 2.0\%$ )	4.9 ( $\pm 8.5\%$ )
<i>Am-243 (t=0yrs)</i>	256.0 ( $\pm 1.0\%$ )	10.1 ( $\pm 3.3\%$ )
<i>Am-243 (t=2 yrs)</i>	1948 ( $\pm 0.7\%$ )	457.5 ( $\pm 0.7\%$ )
<i>Cm-243</i>	3.1E+5 ( $\pm 0.7\%$ )	74300 ( $\pm 0.7\%$ )
<i>Cm-244</i>	5047 ( $\pm 5.2\%$ )	242.6 ( $\pm 9.5\%$ )

The doses are generally for time zero after “fabrication”. For some of the nuclides, it is recognized that the highest activities are obtained after this initial state. Therefore, the values for two years after fabrication are provided for those nuclides. Specifically, Np-237 produces Pa-233, which has the same activity at this time. Similarly, Pu-241 decays to produce mainly Am-241 and some U-237. Am-243  $\alpha$ -decays to give Np-239.

## APPENDIX H

### Radiotoxicity of $\text{UO}_2$

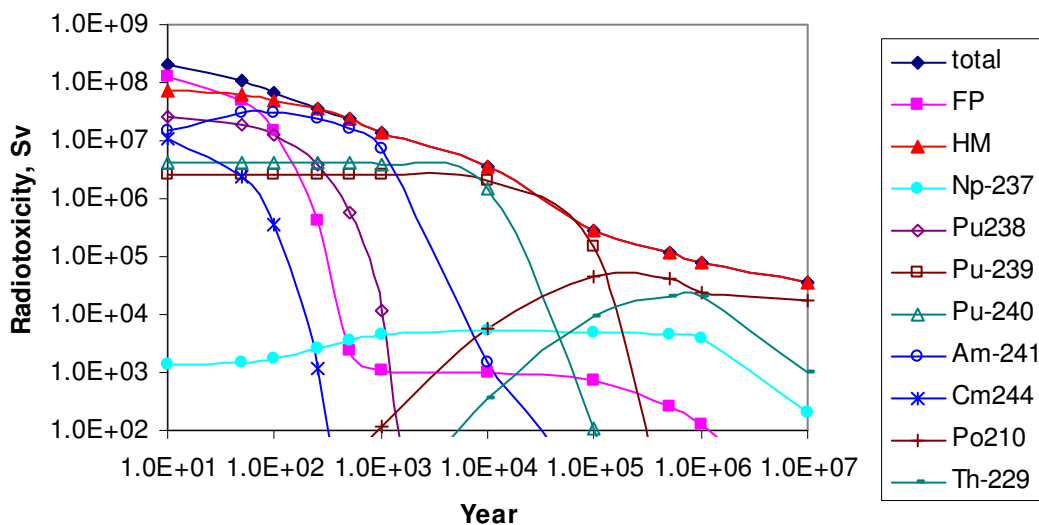


Fig. H.1. Leading Contributors to the Radiotoxicity of the Repository in  $\text{UO}_2$  Case.

### Radiotoxicity of CORAIL-Pu

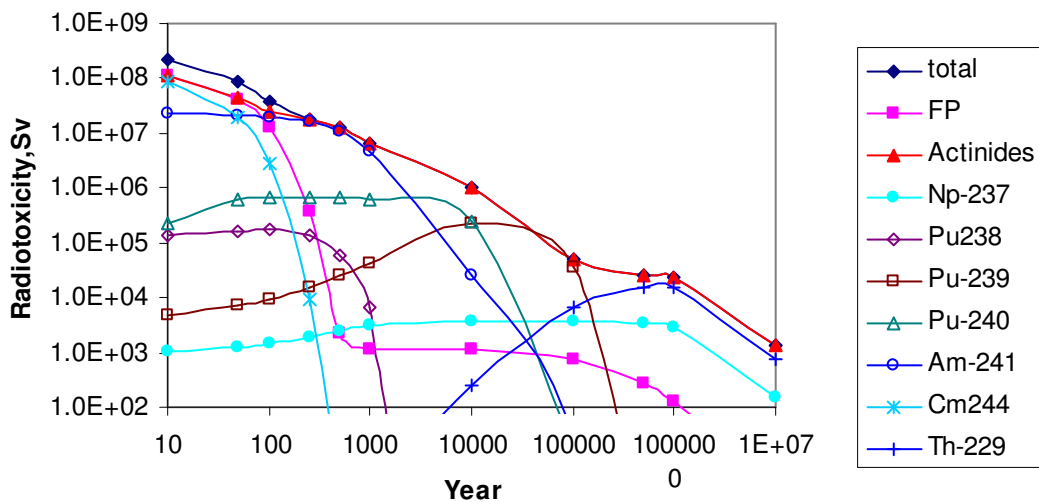
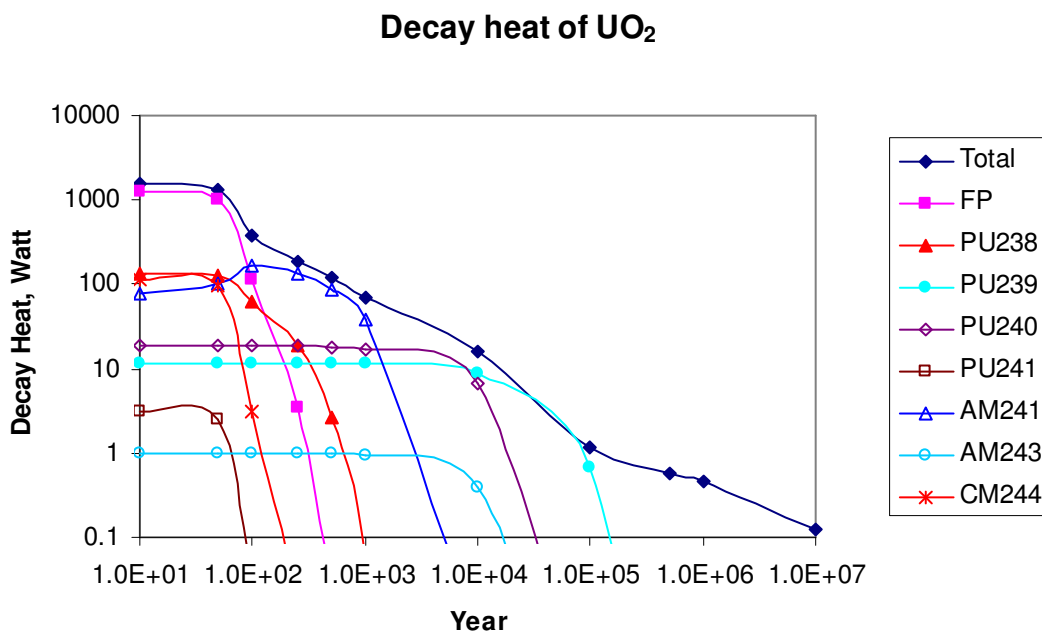
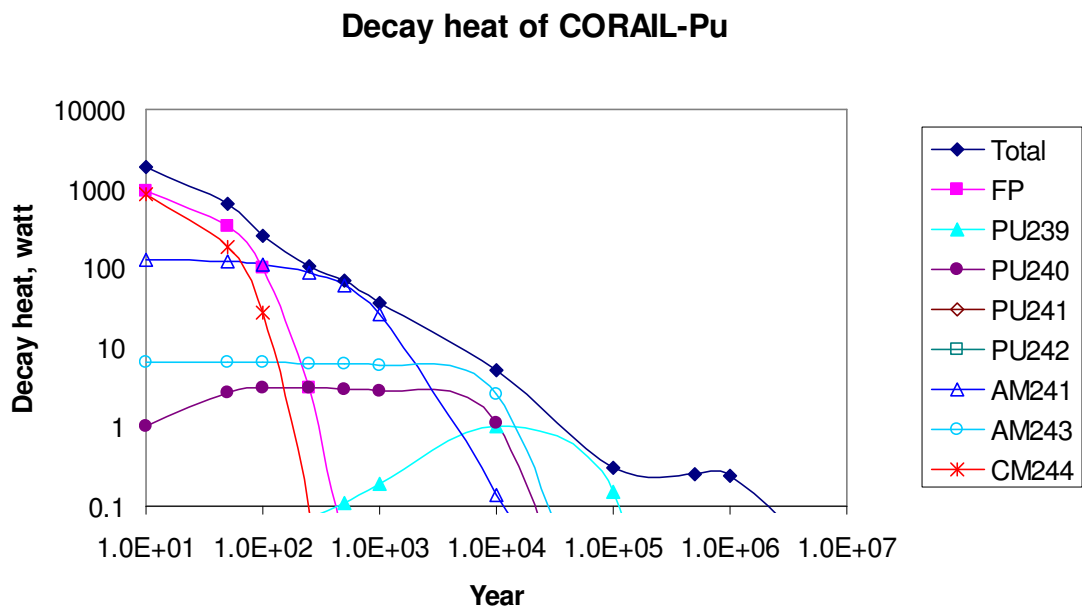


Fig. H.2. Leading Contributors to the Radiotoxicity of the Repository in CORAIL Case.



**Fig. H.3. Leading Contributors to Spent Fuel Decay Heat for UO<sub>2</sub> Case.**



**Fig. H.4. Leading Contributors to Spent Fuel Decay Heat for CORAIL-Pu Case.**

## APPENDIX I

### Typical Fuel Fabrication Gamma Dose Rates for MOX Fuel Containing 4.3wt% PuO<sub>2</sub> (mrem/hr)

Manufacturing Stage	PuO <sub>2</sub>		MOX Pellets		900 MWe PWR all-Pu Assembly	
Days since Am separation	200	600	200	600	200	600
Surface dose (unshielded)	6000	18000	1400	2300	50	80
Surface dose (64-mm lead shield)	15	15	5	5	3	3
Distance of 0.61 m from surface (unshielded)	-	-	-	-	5	8

**Source:** Bairiot and Vandenberg, *Use of MOX Fuels*, Technical Reports Series No. 305, IAEA, Vienna (1989) 65-95; page 73.

## APPENDIX J

### Comparison of Reactivity Coefficients [2,3]

			UO <sub>2</sub>	CORAIL-Pu	CORAIL-TRU
Cycle <sup>a)</sup>			-	7	7
Reactivity Coefficients	Boron worth (pcm/ppm)	Charge	-6.7	-5.3	-4.9
		Discharge	-9.4	-4.2	-2.7
	FTC (pcm/K)	Charge	-2.2	-2.4	-2.4
		Discharge	-3.6	-4.4	-4.3
	MTC (pcm/K)	Charge	-3	-16	-18
		Discharge	-72	-66	-45
	Void (pcm/% void)	Charge	-259	-243	-243
		Discharge	-693	-527	-401

a) Fuel compositions used for evaluating the reactivity coefficients were obtained from WIMS8 calculations.

## APPENDIX K

### TRANSEQM-Calculated Masses for Charge Stages.

	UOX	MOX	CORAIL- Pu	CORAIL-TRU						
Cycle	-	-	7	2	3	4	5	6	7	Equilib.
U enrichment, %	4.0		4.57	4.85	4.98	5.02	5.03	5.04	5.04	5.12
TRU content, %		9.4	8.18	7.48	8.39	9.37	10.31	11.16	11.93	20.2
Fissile, %	4.0	63.63	46.84	49.37	45.23	42.72	41.20	40.13	39.31	32.83
U234		0.051	0.064							
U235	4.0	0.003	0.002							
U236		0.006	0.006							
U238	96.0									
Np237				1.763	2.403	2.570	2.558	2.493	2.419	1.82
Np239				0.000	0.000	0.000	0.000	0.000	0.000	0.00
Pu238		3.134	3.895	2.810	5.043	6.629	7.555	8.119	8.495	10.20
Pu239		56.346	36.056	38.399	34.607	32.565	31.333	30.407	29.660	24.40
Pu240		26.610	26.967	26.885	23.469	21.984	21.318	21.072	21.002	21.67
Pu241		7.283	10.784	10.804	10.390	9.617	9.127	8.816	8.597	7.13
Pu242		5.829	21.136	10.190	12.147	13.359	14.048	14.496	14.828	16.93
Am241		0.738	1.092	5.271	5.827	5.707	5.601	5.562	5.562	5.91
Am242m				0.015	0.031	0.036	0.038	0.041	0.043	0.07
Am243				2.406	3.351	3.730	3.921	4.031	4.104	4.59
Cm242				0.000	0.000	0.000	0.000	0.000	0.000	0.00
Cm243				0.009	0.026	0.030	0.030	0.030	0.029	0.03
Cm244				1.059	2.208	2.946	3.369	3.606	3.741	3.90
Cm245				0.140	0.424	0.637	0.778	0.869	0.929	1.14
Cm246					0.073	0.180	0.302	0.423	0.537	1.74
Cm247					0.000	0.007	0.015	0.024	0.034	0.15
Cm248					0.000	0.002	0.005	0.011	0.019	0.29
Bk249					0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	3.00E-05
Cf249					0.00E+00	6.00E-05	2.20E-04	5.00E-04	9.10E-04	1.91E-02
Cf250					0.00E+00	1.00E-05	5.00E-05	1.00E-04	1.80E-04	3.48E-03
Cf251					0.00E+00	1.00E-05	5.00E-05	1.20E-04	2.10E-04	4.95E-03
Cf252					0.00E+00	0.00E+00	1.00E-05	2.00E-05	4.00E-05	9.40E-04
Cf253					0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+0
Es253					0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+0

NOTE: For the MOX and CORAIL cases, the vectors are for the TRU content of MOX fuel pin.